Gross N transformation rates and related N2O emissions in Chinese and UK agricultural soils

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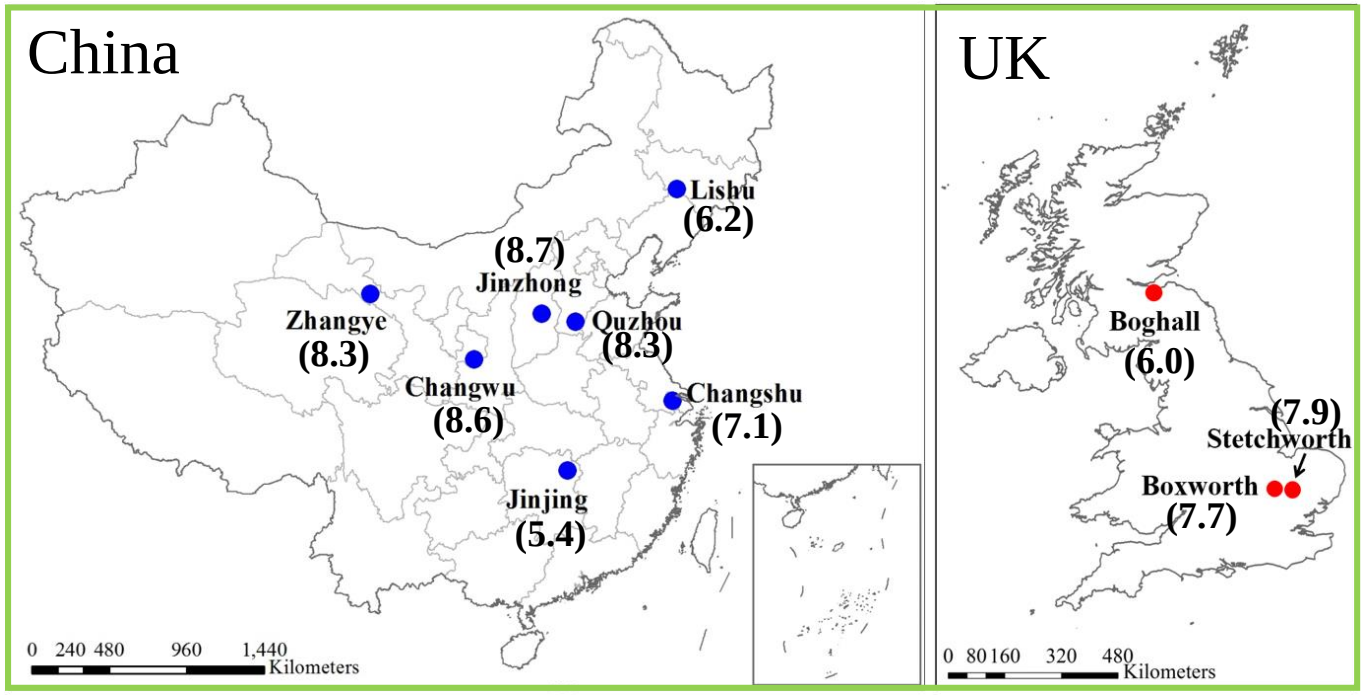
Published in:
Science of the Total Environment

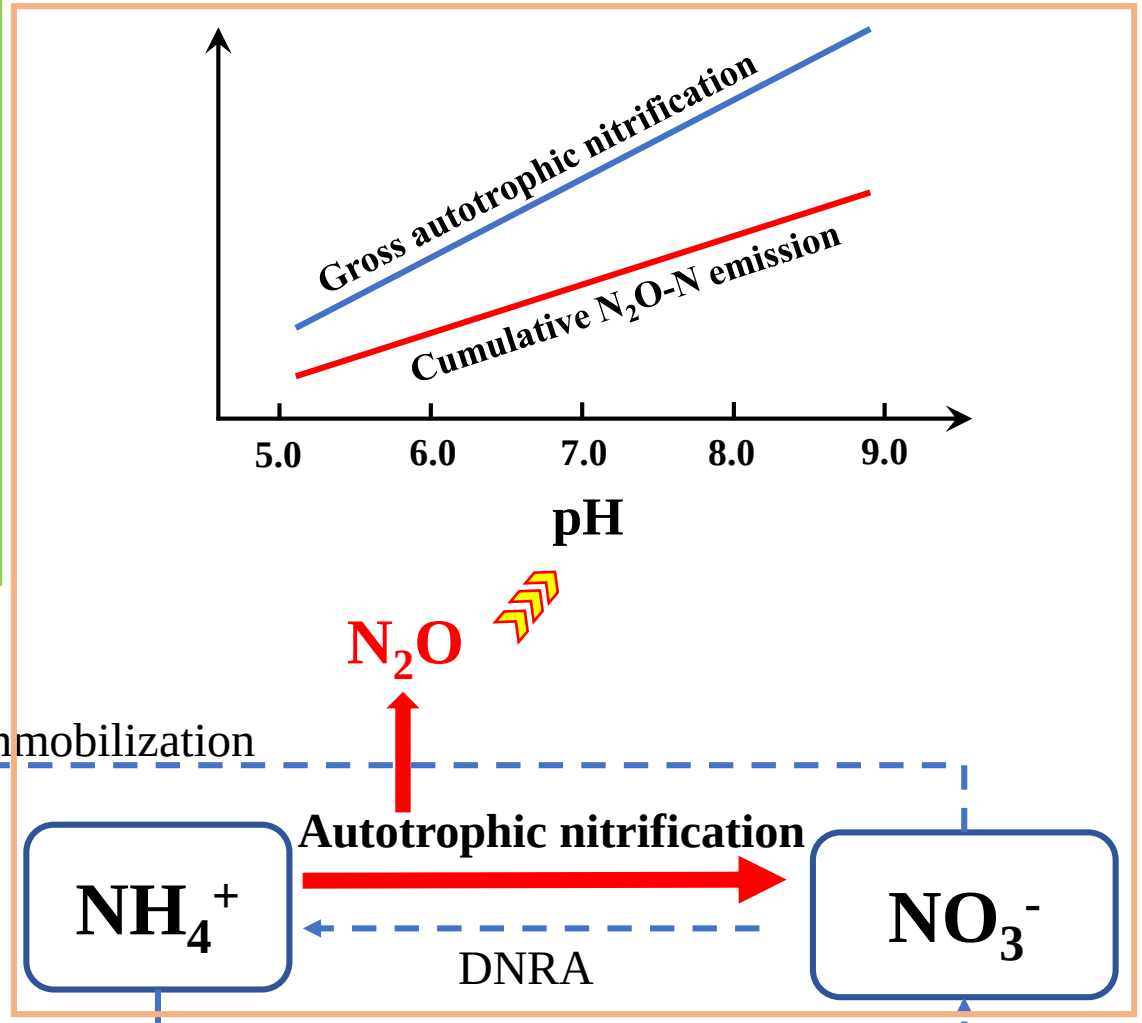
DOI:
10.1016/j.scitotenv.2019.02.241

Print publication: 20/05/2019

Document Version
Peer reviewed version



Note: values in the brackets are soil pH



Highlights

We examined gross N transformations in Chinese and UK agricultural soils.

Autotrophic nitrification and mineralization were the key N transformation processes.

Autotrophic nitrification was important for N_2O emission under aerobic conditions.

pH was the primary factor controlling autotrophic nitrification and N_2O emissions.

Gross N transformation rates were not related to the origin of soil sampling sites.

Gross N transformation rate and related N_2O emission in seven Chinese agricultural soils as comparison with three UK agricultural soils

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Submitted to: Science of the Total Environment

Type of contribution: Research paper

Date of preparation: December 21, 2018

Number of text pages: 39

Number of figures: 6

Number of tables: 2

Gross N transformation rates and related N_2O emissions in Chinese and UK agricultural soils

Abstract

The properties of agricultural soils in various regions of the world are variable and can have a significant but poorly understood impact on soil nitrogen (N) transformations and nitrous oxide (N_2O) emissions. For this reason, we undertook a study of gross N transformations and related N_2O emissions in contrasting agricultural soils from China and the UK. Seven Chinese and three UK agricultural soils were collected for study using a ^{15}N tracing approach. The soil pH ranged from 5.4 to 8.7, with three acidic soils collected from Jinjing, Lishu and Boghall; one neutral soil collected from Changshu, and the other six alkaline soils collected from Quzhou, Zhangye, Changwu, Jinzhong, Boxworth and Stetchworth. Our results showed that the main N transformation processes were oxidation of ammonium (NH_4^+) to nitrate (NO_3^-) (O_{NH4}), and mineralization of organic N to NH_4^+. The gross autotrophic nitrification rates calculated in the three acidic soils were between 0.25 and 4.15 mg N $kg^{-1}d^{-1}$, which were significantly lower ($p<0.05$) than those in the remaining neutral and alkaline soils ranging from 6.94 to 14.43 mg N $kg^{-1}d^{-1}$. Generally, soil pH was positively correlated ($p<0.001$) with gross autotrophic nitrification rate and cumulative N_2O emissions, indicating that soil pH was an important factor regulating autotrophic nitrification and N_2O emissions. There was also a significant positive correlation between the gross autotrophic nitrification rate and cumulative N_2O emissions, highlighting the importance of this process for producing N_2O emissions in

these agricultural soils under aerobic conditions. Gross NH_4^+ immobilization rates were very low in most soils except for the Jinjing soil with the lowest pH. In conclusion, the gross autotrophic nitrification rates and related N_2O emissions were controlled by soil pH irrespectively of the soil's origin in these agricultural soils.

Keywords: Chinese agricultural soils; UK agricultural soils; gross N transformation; N_2O emissions; ^{15}N tracing approach; soil pH

1. Introduction

Soil nitrogen (N) transformations (including processes and rates) play a crucial role in regulating soil N availability and N losses (Galloway et al., 2008). While soil N mineralization provides an important source of plant-available N (Wang et al., 2015), the reverse process of immobilization transfers mineral N into the organic N pools and is an important N sequestration mechanism in soils, mediated by microbial activity linked to the humification processes (Ponge, 2013). Thus, soil N mineralization-immobilization turnover (MIT) governs soil N availability (Murphy et al., 2003; Huygens et al., 2007), which is regulated by soil pH, organic carbon (C), N C/N ratios and microbial community when soil temperature and moisture conditions do not limit microbial activity (Booth et al., 2005; Wang et al., 2015).

Nitrification including autotrophic and heterotrophic pathways produces nitrate (NO_3^-) and regulates the ratio of ammonium (NH_4^+) to NO_3^- in soils. It also controls N loss through leaching or as gaseous emissions e.g. nitrous oxide (N_2O) (Zhang et al., 2018). Autotrophic nitrification is driven by chemo-autotrophic nitrifiers which oxidize ammonia (NH_3) to NO_3^- and gain energy from this process, while heterotrophic nitrification is carried out by heterotrophic nitrifiers or fungi, which can oxidize either NH_4^+ or organic N to form NO_3^-, and utilize organic C for their energy (Zhang et al., 2014; Liu et al., 2015). Because autotrophic nitrifiers are sensitive to low pH, it is thought that heterotrophic nitrification may play a predominant role in NO_3^- production in acidic soils (Huygens et al., 2007; Zhang et al., 2013a).

52 The N transformation processes that are capable of producing N_2O such as
53 autotrophic nitrification, heterotrophic nitrification, nitrifier-denitrification, coupled
54 nitrification-denitrification, denitrification and dissimilatory nitrate reduction to
55 ammonia (DNRA) can occur simultaneously in soils. These processes are regulated
56 by a range of soil properties, such as soil pH, texture, C and N content,
57 aerobic-anaerobic micro-sites and the microbial community structure which is
58 influenced by environmental factors (temperature, water content and oxygen)
59 (Butterbach-Bahl et al., 2013; Müller et al., 2014). Rates of N cycling are strongly
60 influenced by the magnitude of pools of soil NH_4^+, organic N and NO_3^- which in turn
61 are strongly affected by N additions in the form of manufactured N fertilizer or
62 manure (Snyder et al., 2009). However, under aerobic conditions, N_2O production
63 from denitrification in agricultural soils with a high pH and low organic C content is
64 minor relative to autotrophic nitrification. For instance, N_2O emissions occur mainly
65 as a consequence of the ammonia oxidation process in intensively managed low
66 organic C (5-10 g kg^{-1}) calcareous (with a pH of between 7.2 - 8.5) agricultural soils
67 in the North China Plain (Wan et al., 2009; Ju and Zhang, 2017). By contrast, in
68 agricultural soils in the UK with high organic C (about 23 to 40 g kg^{-1}) (King et al.,
69 2005), high water contents together with relatively low soil pH (ranging from 5.0 to
70 7.5; Goulding, 2016), denitrification is a major contributor to overall N_2O emissions
71 (Clark et al., 2012).
72 The properties of agricultural soils in China and the UK are highly variable and
73 depend on parent material, climate, topography, vegetation and time, as well as land

74 use, and agricultural management practices (e.g. crop type, tillage, fertilization and
75 irrigation, etc.). For the purpose of this study we chose agricultural soils from
76 different parts of China and the UK that were representative of the agricultural
77 landscapes in these countries and therefore relevant to the research questions being
78 addressed by our study. The upland agricultural soils of Northern China are located in
79 a region to the north of a line connecting the Kunlun Mountains, the Qinling
80 Mountains and the Huaihe River. They have a temperate monsoon or continental
81 climate with dry cold winters and wet hot summers (Ju and Zhang, 2017). Based on
82 differences in soil properties, precipitation and agricultural management practices,
83 three major sub-regions, namely the Northeast Plain, the North China Plain and
84 Northwest China (including Loess Plateau), can be identified in this region of China
85 (Liu and Chen, 2005). The soils are characterized by high organic C contents ranging
86 from 8 to 44 g kg^{-1} (Liu et al., 2006) and relatively low pH (5.5-7.2) (Shangguan et al.,
87 2013) in the Northeast Plain, but low organic C contents (5-10 g kg^{-1}) (Liao et al.,
88 2015) and high pH (7.2-9.8) (Shangguan et al, 2013) in the North China Plain and
89 Loess Plateau. However, the soils in the South China region (mainly paddy soils) are
90 located in a subtropical monsoon climate zone and are characterized by a low pH
91 ranging from 4.5 to 5.5 (with a few exceptions that are characterized by neutral pH)
92 (Shangguan et al., 2013). They are also highly weathered and have a poor buffering
93 capacity (Zhao et al., 2007). In addition, Chinese agricultural soils are often
94 intensively managed with frequent tillage, high N fertilization rates and a lack of

95 manure and crop residue inputs, which leads to soil acidification, low soil fertility and
96 reduced N retention (Ju et al., 2009; Guo et al., 2010).

97 Tilled agricultural soils in the UK are mainly located in the East of Scotland, and
98 the East and South of England (Brown and Shipley, 1982; Avery, 1990; Davies et al.,
99 1996), and are characterized by a temperate maritime climate, where the seasonal
100 temperature change is moderated by relatively cool summers and temperate winters.
101 The agricultural soils in the East of Scotland tend to have more organic C and be more
102 acidic than soils in the rest of the UK, with a texture that is predominantly sandy-loam
103 to loam, and often exhibit poor drainage (Hay et al., 2000). In the East and South of
104 England the proportion of alkaline soils is higher than in Scotland, and although
105 textures are typically heavier (clays and loams dominate large areas) (Dobbie and
106 Smith, 2003), the organic C concentrations are generally lower overall due to higher
107 temperatures and generally lower rainfall.

108 Previous research in the UK has shown that the delayed uptake of applied N in
109 wheat was associated with temporary net immobilization by soil microbes, and thus
110 could not temporarily supply mineral N in the rapid growth period of the plant (King
111 et al., 2001; Silgram and Chambers, 2002; Murphy et al., 2007). The low C and
112 calcareous soils of the North China Plain, by contrast, have a low potential for
113 immobilization, high rates of nitrate leaching and N_2O emissions are associated with
114 excessive net mineralization and nitrification (Ju and Zhang, 2017). Our hypothesis in
115 approaching this work was that the relatively high organic C contents associated with
116 UK soils would favour N immobilization. However, to date, differences in the N

117 transformation characteristics and related N_2O emissions from contrasting agricultural
118 soils in China and the UK, have not been compared. Such knowledge is a prerequisite
119 to evaluate novel techniques which aim to manipulate soil MIT, nitrification, and
120 denitrification rates to enhance the recovery of applied N and to provide information
121 on cropping systems, which can in turn help to optimize N fertilization and reduce N
122 losses.

123 Therefore, the objectives of this study were: 1) to quantify the gross N
124 transformation rates and N_2O emissions in a typical selection of Chinese and UK
125 agricultural soils; 2) to study factors controlling the N transformation process
126 responsible for N_2O emissions from these soils. Seven Chinese and three UK
127 agricultural soils were collected to study the above objectives.

128 **2. Materials and methods**

129 2.1 Site description and soil samples collection

130 Soils were sampled from seven typical Chinese agricultural sites and three UK
131 agricultural sites in 2016 (Fig. S1), which refers to dominant soil types and crop
132 systems in the main agricultural regions of China and UK. At the sampling sites
133 typical management practices for the local crops were adopted. Some important soil
134 and climatic properties, land use and crops for each site are summarized in Table 1.

135 Soil samples were collected after crop harvest between October and December
136 2016 from the plough layer (0-20 cm) at each site. Ten to fifteen soil cores were
137 randomly taken from each field site (approximately 400 m^2) in a zigzag sampling
138 pattern using stainless steel augers (5 cm diameter) and mixed to produce one

139 composite sample (approximately 3 kg). After collection, soils were stored in a cool
140 box, before being transferred to the laboratory where they were sieved to 2 mm. Roots
141 and any visible organic substrates were removed by tweezers to reduce the
142 heterogeneity. Each soil sample was divided into two parts, and approximately 400 g
143 of the soil was air-dried for analysis of soil properties, and the remaining soil was
144 stored at 4.0 ℃ for the incubation experiment.

145 2.2 ^{15}N tracing incubation experiment

146 Gross N transformation rates were quantified using a ^{15}N tracing approach
147 (Kirkham and Bartholomew, 1954; Müller et al., 2007; Zhang et al., 2012a; Wang et
148 al., 2017). For each soil, there were two ^{15}N-labelled treatments: ^{15}N labelled NH_4^+
149 ($^{15}NH_4NO_3$ at 9.76 atom % ^{15}N excess) and ^{15}N labelled NO_3^- ($NH_4{}^{15}NO_3$ at 9.74
150 atom % ^{15}N excess). For each soil, 24 Erlenmeyer flasks were prepared (2 treatments,
151 3 replicates, and 4 extraction times) with 20 g (oven-dry basis) of fresh soil, which
152 were pre-incubated for 24 h at about 50% water holding capacity (WHC) and at 20℃.
153 Here the 50% of WHC referred to water holding capacity of the sieved soil, which
154 was higher than the WHC measured of well-structured soil *in situ,* but was an
155 appropriate water potential to promote microbial activity under aerobic conditions.
156 Two milliliters of $^{15}NH_4NO_3$ or $NH_4{}^{15}NO_3$ solution were added to each Erlenmeyer
157 flask at a rate of 20 mg NH_4^+-N kg^{-1} or 20 mg NO_3^--N kg^{-1} (equivalent to 40 mg N
158 kg^{-1}), respectively. The soils were adjusted to 60% water holding capacity (WHC) and
159 sealed with parafilm® with four pinholes to allow aeration, and incubated in the dark
160 for 48 h at 20℃. This represents a routine procedure for these kinds of studies (Müller

et al., 2007; Zhang et al., 2012a), and allows the investigation of fundamental processes and rates of soil N transformation under conditions conducive to high rates of microbial activity. The soil N transformations measured at the incubation temperature of 20℃ in this study would be close to conditions experienced at the range of sampling sites. After incubation, soils were extracted with 2M KCl at 0.5, 12, 24 and 48 h after labelling to determine the concentrations and isotopic compositions of NH_4^+ and NO_3^-.

Additionally, another 6 flasks of each soil (two ^{15}N-labelled treatments and three replicates) were prepared for measuring N_2O fluxes, which were treated in the same way. Gas samples were collected at 12, 24 and 48 h after N application. Before sampling, the flasks were flushed with ambient air using a multiport vacuum manifold. The above procedure was repeated three times (each time for about 10 s). Thereafter, the flasks were immediately sealed for 4 h using a silicone sealant and gas samples were collected using a syringe fitted with a three-way stopcock and transferred to 18.5 ml pre-evacuated vial, to determine the N_2O concentration (using an Agilent 7890 GC, USA). The N_2O flux was calculated as:

$$F_{N2O} = \{[M \times C \times Vg \times 273]/[W \times (273+T) \times 22.4]/t\} \times (28/44) \quad (1)$$

where F_{N2O} (μg N kg^{-1} h^{-1}) is the N_2O flux; M (g mol^{-1}) is the molar mass of N_2O; C (ppm) is the concentration of N_2O; Vg (L) is the volume at the top of the flask; W (kg) is the dry soil weight; T (℃) is mean air temperature during incubation; 22.4 (L mol^{-1}) is the molecular volume at 1,013 hPa and 273 K; t (h) is the time of flask closure; 28/44 is used for unit conversion. The fluxes of N_2O measured on each sampling date

were used to determine cumulative N_2O emissions by the trapezoidal integration method.

2.3 Chemical analysis

Soil NH_4^+ and NO_3^- were extracted with 2 M KCl at a soil / solution ratio of 1:5 (v/v) on a mechanical shaker for 1 h at 300 rpm at 20 °C. The extract was passed through filter papers (Whatman No. 42 Cat No. 1442-055) and the concentrations of NH_4^+ and NO_3^- were analyzed using a continuous-flow analyzer (SA1000, Skalar, the Netherlands). The isotopic composition of NH_4^+ and NO_3^- was determined using the modified micro-diffusion method (Zhang et al., 2017) and measured using a Delta V plus isotope mass spectrometer (Thermo Fisher Scientrific, Delta V plus, Germany). Briefly, a portion of the extract was mixed with MgO to release NH_3 from the NH_4^+ pool. Devarda's alloy was then added to the flask to reduce NO_3^- to NH_4^+, and then to NH_3. Liberated NH_3 was trapped using filter paper, which was acidified with 1 M oxalic acid. After diffusion, filters were transferred to a free-ammonia environment for drying, then dried filter papers were transferred to a tin capsule and wrapped to enable the enrichment of ^{15}N to be analysed. Soil pH was determined at a soil to water ratio of 1:2.5 (w/v) using a pH meter (Mettler Toledo, Switzerland). Soil organic carbon (SOC) and total N was determined with a C/N element analyser (Sample preparation system, Europa EA-GSL).

2.4 ^{15}N tracing model

The *Ntrace* model was used to quantify gross N transformations rates (Müller et al., 2007). The measured concentrations and ^{15}N enrichment values (mean ± standard

deviation) of NH_4^+ and NO_3^- from the triplicate measurements in the two ^{15}N treatments were supplied to the model, and gross N transformation rates were calculated by simultaneously optimizing the kinetic parameters for each individual process by minimizing the misfit between modelled and observed concentrations of NH_4^+ and NO_3^- and their respective ^{15}N enrichments. To obtain the most appropriate model, several model modifications, which vary in the number of considered N pools and processes, and kinetic settings, were tested (Rütting and Müller, 2008). The most appropriate model was guided by the Aikaike's information criterion (AIC), selecting the minimum AIC value (Cox et al., 2006; Rütting and Müller, 2007; Wang et al., 2016). In addition, the determination coefficient (R^2) can also be used to verify the model; if R^2>0.80, the modelled result can be accepted (Quinn and Keouogh, 2002). The numerical optimization model based on Markov Chain Monte Carlo Metropolis algorithm, can provide reliable results for a large number of parameters. For more detailed information on the model development and parameter optimization see Müller et al. (2007).

Ten simultaneously occurring transformations were quantified (Müller et al., 2007; McGeough et al., 2016): 1) M_{Nrec}, mineralization of recalcitrant organic-N to NH_4^+; 2) M_{Nlab}, mineralization of labile organic-N to NH_4^+; 3) I_{NH4_Nlab}, immobilization of NH_4^+ to labile organic N; 4) I_{NH4_Nrec}, immobilization of NH_4^+ to recalcitrant organic N; 5) R_{NH4ads}, release of adsorbed NH_4^+; 6) A_{NH4}, absorption of NH_4^+ on cation exchange sites; 7) O_{NH4}, oxidation of NH_4^+ to NO_3^-; 8) O_{rec}, oxidation of recalcitrant organic N to NO_3^-; 9) I_{NO3}, immobilization of NO_3^- to recalcitrant

organic-N; and 10) D_{NO3}, dissimilatory NO_3^- reduction to NH_4^+. Gross transformation rates were calculated by zero- or first-order kinetics. In this model, labile organic N was assumed to be the maro-organic matter fraction and accounted for 1% of total organic N according to Rütting and Müller (2008) and Chen et al. (2015). The optimization procedure resulted in a probability density function for each parameter, from which averages and standard deviations were calculated (Müller et al., 2007). Each analysis run was carried out with three parallel sequences. Based on the kinetic setting and the final parameters, average N transformation rates were calculated over the initial 24 h incubation period (because the concentrations and abundances of NH_4^+ could not be reliably detected at 48 h in most soil samples) and expressed in units of mg N kg^{-1} dry soil d^{-1} (Table 2).

Net mineralization rates were calculated from the differences in NH_4^+ plus NO_3^- concentration between two sampling dates divided by the number of days in the sampling intervals, while net nitrification rate was calculated from the difference in NO_3^- concentration (Table S1).

2.5 Statistical analyses

For gross rates of N transformation, most statistical tests are inappropriate for the comparison of parameter results, because of the large number of iterations of the ^{15}N tracing model (Yoccoz, 1991). Thus, parameter results based on the comparisons of standard deviations and the 95% confidence intervals were used to distinguish three cases: a) standard deviation overlap: the parameters are not different; b) standard deviations do not overlap but 95% confidence intervals overlap, the parameters are

249 not significantly different but show a clear tendency to be different; c) 95%
250 confidence intervals do not overlap, parameters are significantly different (Müller et
251 al., 2011). A one-way analysis of variance (ANOVA) with a least significant
252 difference test (p<0.05) was used to compare the cumulative emissions of N_2O in the
253 ten soils. Correlation and linear or nonlinear regression analyses were used to test
254 relationships between soil pH and total gross mineralization rate, autotrophic
255 nitrification rates and cumulative N_2O emissions, cumulative N_2O emissions and total
256 gross mineralization and autotrophic nitrification rate, and the relationship between
257 total gross mineralization and gross autotrophic nitrification. All the statistical
258 analyses were performed using SPSS software package 17.0 for windows (Inc., USA)
259 and correlations were analyzed using SigmaPlot 12.5 (Systat Software Inc., Erkrath,
260 Germany).
261

262 **3. Results**

263 3.1 Soil properties

264 Soil properties showed considerable variation in the ten soils used for this study
265 (Table 1). The soil pH ranged from 5.4 to 8.7, with pH values < 6.5 in the soils
266 collected from Jinjing, Lishu and Boghall (acidic soils); one soil collected from
267 Changshu had a pH value of 7.1 (neutral soil), while all the remaining soils had pH
268 values ≥7.6 (alkaline soils). Soil organic C ranged from 21.9 to 27.4 g kg^{-1} in soils
269 from Jinjing, Changshu, Boghall, Boxworth and Stetchworth and these were
270 significantly higher (p<0.05) than the other soils which ranged from 8.1 to 17.8 g kg^{-1}
271 (Lishu, Quzhou, Zhangye, Changwu and Jinzhong). The SOC contents of all three UK
272 soils (21.9 to 27.4 g kg^{-1}) and two paddy soils from Jinjing and Changshu (25 to 27.1
273 g kg^{-1}) was relatively high.

274 High SOC contents, were associated with high N contents in the two Chinese
275 paddy soils and the three UK soils ranging from 2.0 to 3.0 g N kg^{-1}, which was
276 significantly higher (p<0.05) than the rest (ranging from 0.7 to 1.7 g N kg^{-1}). The C/N
277 ratio of all soils varied from 9.0 to 12.3, which were typical for such tilled agricultural
278 soils. The inorganic N was dominated by NO_3^- in all soils, ranging from 9.02 to 62.36
279 mg N kg^{-1}, and the minimum and maximum values were measured in the soils from
280 Changshu and Boghall, respectively. The NH_4^+-N concentrations in soils from Jinjing
281 and Boxworth were 8.03 mg N kg^{-1} and 4.70 mg N kg^{-1}, respectively, and were
282 substantially higher (p<0.01) than the others, which ranged from 0.03 to 1.41 mg N

283 kg^{-1}. In addition, the NH_4^+-N/NO_3^--N ratio of all soils varied from 0 to 0.52, and the
284 maximum value were observed in Jinjing soil.

285 3.2 Concentrations and enrichment of NH_4^+ and NO_3^- pools

286 To reveal the relationship between NH_4^+ consumption and NO_3^- production
287 within different incubation periods, we calculated the net change of NH_4^+ (ΔNH_4^+)
288 and NO_3^- (ΔNO_3^-) concentrations (Table S1, Fig. S2), and found that a decline in
289 NH_4^+ concentration generally corresponded well with an increase of NO_3^-
290 concentration in all soils within different incubation periods, but the rate of NH_4^+
291 oxidation to NO_3^- during different incubation periods varied. During the period
292 between 0-0.5 h, it was found that $\Delta NH_4^+ < \Delta NO_3^-$ in all Chinese soils, but this was
293 reversed in the three UK soils (Fig. S2 a). However, during the periods 0.5-12 h,
294 12-24 h and 24-48 h, the ΔNH_4^+ was almost equal to the ΔNO_3^- in all soils. The value
295 of ΔNO_3^- was slightly higher than ΔNH_4^+ in most cases between 0.5-24 h and 0.5-48 h
296 of incubation, indicating a low net mineralization of soil organic N compared to a net
297 oxidation of NH_4^+.

298 The dynamics of $NH_4^+$and NO_3^- concentrations in the ten soils from $^{15}NH_4NO_3$
299 and $NH_4{}^{15}NO_3$ treatments were almost the same within the 48 h incubation period
300 (Fig. 1). There was a gradual decline in NH_4^+ concentrations, while NO_3^-
301 concentrations gradually increased during the incubation of the neutral and alkaline
302 soils (Fig. 1). By contrast, there was relatively little change in the NH_4^+ and NO_3^-
303 pools in the acidic soils, indicating a weaker ammonia oxidation ability in the acidic
304 soils. Model simulations of $NH_4^+$and NO_3^- dynamics corresponded well with

measurements within the 24 h incubation period. Model evaluations were only possible for the first 24 hours due to fast N transformation processes depleting the mineral N pools so that the ^{15}N excess of the NH_4^+ and NO_3^- could not be determined (see next section) (Table 2).

In the labelled $^{15}NH_4NO_3$ treatment, the atom % ^{15}N excess of NH_4^+ pool decreased, while the atom % ^{15}N excess of NO_3^- pool increased during the incubation in the neutral and alkaline soils, but not in the acidic soils (Fig. 2). This corresponded well with the above changes of NH_4^+ and NO_3^- concentrations in different soil situations, showing a high NH_4^+ oxidation activity in soils with a pH >7.0. The enrichment of NH_4^+ pools after 0.5 h incubation of the labelled $^{15}NH_4NO_3$ treatment ranged between 7.87-8.15 in all soils (i.e. close to the applied 9.76 atom% ^{15}N excess), except for soils from Jinjing (5.42 atom% ^{15}N excess) and Boxworth (7.65 atom% ^{15}N excess) indicating that the ^{15}N dilution was low (Fig. S3 a). Simulated and observed isotopic enrichments corresponded well within the 24 h incubation. The enrichment of the NH_4^+ pool in the labelled $^{15}NH_4NO_3$ treatment decreased sharply between 24 and 48 h in the neutral and alkaline soils but not noticeably in the acidic soils (Fig. 2 a-j). These situations could be also illustrated by the significant correlation between the isotopic enrichment of NH_4^+ pools in the 24-48 h period and the NH_4^+ concentration at 24 h, in which small changes in the acidic soils from Jingjing, Lishu and Boghall corresponded with relatively high NH_4^+ concentrations after 24 h (Fig. S4).

In the labelled $NH_4{}^{15}NO_3$ treatment, the enrichment of the NO_3^- pool decreased with the increasing incubation time in the neutral and alkaline soils, but not

noticeably in the acidic soils (Fig. 2 a'-j'). The enrichment of the NH_4^+ pool was extremely low (ranging between 0.01-0.03 atom%) over the whole incubation period, indicating that the NH_4^+ transformed from labelled NO_3^- pool was negligible. The enrichment of the NO_3^- pool after 0.5 h in the labelled $NH_4{}^{15}NO_3$ treatment was in the range of 2.25-5.58 atom% in all soils compared to the initial labelled $NH_4{}^{15}NO_3$ enrichment of 9.74 atom% ^{15}N excess of NO_3^- pool. Due to the relatively high initial NO_3^- concentrations in all soils a ^{15}N dilution effect was observable. We also observed a negative relationship ($p<0.01$) between initial NO_3^- concentration and atom % ^{15}N excess after 0.5 h in the NO_3^- pool in the labelled $NH_4{}^{15}NO_3$ treatment (Fig. S3 b).

3.3 Gross N transformation rates

The main processes studied in these soils were oxidation of NH_4^+ to NO_3^- (O_{NH4}), and mineralization of labile (M_{Nlab}) and recalcitrant (M_{Nrec}) organic N to NH_4^+, except for the soil from Jinjing with the lowest pH (5.4) which was also characterized by relatively high NH_4^+ immobilization to labile (I_{NH4_Nlab}) and recalcitrant (I_{NH4_Nrec}) organic N. Total mineralization rates (M_{tot}) were calculated by: $M_{tot} = M_{Nlab}+M_{Nrec}$.

The maximum total gross mineralization rate was observed in the soil from Jinjing (5.26 mg N $kg^{-1}d^{-1}$), followed by Stetchworth (3.43 mg N $kg^{-1}d^{-1}$), which was significantly higher ($p<0.05$) than that in other soils (1.03-3.24 mg N $kg^{-1}d^{-1}$). Gross NH_4^+ immobilization ($I_{NH4tot} = I_{NH4\_Nlab}+I_{NH4\_Nrec}$) was observed in soil from Jinjing (4.55 mg N $kg^{-1}d^{-1}$), but was negligible in the other soils.

Autotrophic nitrification (O_{NH4}) was responsible for the largest flows of N in

these agricultural soils, except for the Jinjing soil with the lowest soil pH. The gross autotrophic nitrification rates calculated in acidic soils from Jinjing, Lishu and Boghall, were 0.25, 3.05 and 4.15 mg N $kg^{-1}d^{-1}$, respectively, which were significantly lower (p<0.05) than those in the remaining neutral and alkaline soils ranging from 6.94 to 14.43 mg N $kg^{-1}d^{-1}$. This indicates that autotrophic nitrification was favored by high soil pH. Unexpectedly, the heterotrophic nitrification process (O_{Nrec}) was observed in the soil from Lishu, but at a low rate of 0.80 mg N $kg^{-1}d^{-1}$. Compared to NO_3^- production pathways, the NO_3^- consumption pathways, including gross NO_3^- immobilization and dissimilatory NO_3^- reduction to NH_4^+ (DNRA), were negligible for all studied soils (see Table 2).

Within the 0.5-12 h incubation period, gross mineralization was approximately equal to the net mineralization in almost all soils (Fig. S6 a) and was similar in most soils in the 0.5-24 h period, except for soils from Zhangye, Changwu and Jinzhong which had a pH above 8.0 (Fig. S6 b). Within the 0.5-12 h period, the total gross nitrification rates were higher than the net nitrification rates in soils from Lishu and Boghall, while the opposite trend occurred in the other soils (Fig. S6 c). However, the total gross nitrification rates were approximately equal to the net nitrification rates in the soils within the 0.5-24 h incubation period, except for soils from Zhangye, Changwu and Jinzhong with a pH above 8.0 (Fig. S6 d), in which there was lower gross than net nitrification.

3.4 N_2O emissions during incubation

Generally, N_2O fluxes increased and reached a maximum after 24 h of incubation

except for soils from Changshu, Boxworth and Boghall, in which N_2O fluxes were highest at 12 h of incubation, especially in the paddy soil from Changshu (Fig. S5 a). After 24 h, N_2O fluxes decreased gradually and declined to a minimum after 48 h. We calculated the cumulative N_2O emissions during the 24 and 48 h periods, and observed a similar ranking of treatments (Fig. 3 and Fig. S5 b). To be consistent with the period of model simulation, we used the cumulative N_2O emissions within the 24 h incubation to establish the relationships of N_2O emissions with the gross N transformation rate and some soil properties as analyzed in following sections.

The cumulative N_2O emissions (ranged from 0.65 to 1.46 μg N kg^{-1} dry soil) and autotrophic nitrification in three acidic soils (pH<6.5) were both lower than those from other soils with pH>7.0 (cumulative N_2O emissions ranging from 2.29 to 7.12 μg N kg^{-1} dry soil), although the soil from Quzhou was an outlier (with a low cumulative N_2O emission of <2 mg N kg^{-1} dry soil) (Fig. 3). The cumulative N_2O emissions of the remaining six soils were in the order of Changshu > Zhangye > Jinzhong > Boxworth > Stetchworth > Changwu, although the Changshu and Zhangye soils had significantly higher emissions (p<0.01) than the other soils.

3.5 Correlations between gross N rate, N_2O emissions and soil properties

Soil pH was positively correlated (p<0.001) with gross autotrophic nitrification rates and cumulative N_2O emissions (p<0.05) (except for the paddy soil from Changshu which had the highest cumulative N_2O emissions), confirming that soil pH was an important driving factor regulating autotrophic nitrification and N_2O emissions (Fig. 4). In addition, the autotrophic nitrification rate was significantly positively

correlated (p<0.01) with cumulative N_2O emissions (except for the paddy soil from Changshu), but was not correlated with total gross mineralization rates (Fig. 5). This suggests that autotrophic nitrification plays an important role in producing N_2O in these agricultural soils under aerobic conditions. A significant correlation (p<0.05) was found between the total gross mineralization rate and autotrophic nitrification rate in all soils except soil from Jinjing (Fig. 6), suggesting that the availability of NH_4^+ was a critical factor in limiting autotrophic nitrification. The total gross mineralization and autotrophic nitrification rates were not significantly correlated with any other soil properties (Fig. S7).

4. Discussion

4.1 Soil pH, autotrophic nitrification rate and N_2O emissions

Our results showed that autotrophic nitrification (O_{NH4}) was the dominant process of N transformation and that soil pH was the key factor controlling both autotrophic nitrification and N_2O emissions, which is consistent with previous studies (Zhao et al., 2007; Zhang et al., 2013a). The gross autotrophic nitrification rates and cumulative N_2O emissions in the acid soils were much lower than those in neutral or alkaline soils. There was a significant positive correlation between gross autotrophic nitrification and cumulative N_2O emissions (p<0.01), highlighting the importance of the nitrification as a source of N_2O emissions in these agricultural soils under aerobic conditions. However, the soils in the field are not always aerobic, especially in UK. Environments with higher labile C, higher moisture contents and relatively low soil pH can promote denitrification and make a substantial contribution to N_2O emissions

(Dobbie et al., 1999; Dobbie and Smith, 2003). Similar situations also occur in the low C, high pH and well-aerated loamy soils in North China where intense rainfall or irrigation events occur, leading to microoxic or anaerobic conditions, which in turn induce denitrification (Gao et al., 2014; Huang et al., 2014).

The role of pH in controlling overall N_2O emissions is complex, and depends upon the balance of the nitrification process under aerobic conditions and denitrification process under anaerobic conditions, (Butterbach-Bahl et al., 2013). Studies have shown that decreasing soil pH could increase the N_2O/N_2 ratio by denitrification as a consequence of the inhibition of N_2O reductase, resulting in increasing N_2O emissions from denitrification under anaerobic conditions (Šimek and Cooper, 2002; Saggar et al., 2013). However, under aerobic conditions, N_2O is the major product of nitrification, and an increase in the soil pH can then stimulate N_2O production (Signor and Cerri, 2013). This implies that different N_2O mitigation strategies could be adopted under different soil conditions. For example, nitrification inhibitors could be applied to neutral and alkaline soils to slow down the ammonia-oxidizing process and reduce N_2O emissions (Ding et al., 2011; Huang et al., 2014). In circumstances where well drained acidic soils are being used for crop production reducing ammonium supply would be likely to be more effective for reducing N_2O emissions than in wetter soils where denitrification would predominate (Liu et al., 2017). The Changshu paddy soil exhibited higher N_2O emissions compared to other soils, which were attributed to a high pH (7.1) and high SOC & TN providing ideal conditions for producing large N_2O emissions under aerobic conditions

437 (Butterbach-Bahl et al., 2013). The crop rotation in Changshu was paddy rice
438 followed by winter wheat, and the aerobic incubation maintained in the current study
439 could reflect the characteristics of gross N transformation and N_2O emissions in the
440 wheat season.

441 In addition, autotrophic nitrification rates are also controlled by soil pH,
442 primarily through its effects on NH_3 availability (Nugroho et al., 2007; Cheng et al.,
443 2013). Ammonia is the substrate for ammonia-oxidizing microorganisms, and its
444 concentration declines exponentially with decreasing soil pH due to the ionization of
445 NH_3 to NH_4^+, and NH_4^+ uptake requires an active mechanism of transmembrane
446 transport and therefore an energy source which may also be limited at low pH
447 (Kemmitt et al., 2006; Baggs et al., 2010). Thus, low autotrophic nitrification rates in
448 acidic soils are thought to be caused by substrate limitations and a low number of
449 autotrophic nitrifiers (Jiang and Bakken 1999), with pH tolerances varying between
450 different species of ammonia-oxidising bacteria (De Boer et al., 1991). Low soil pH
451 suppresses NH_4^+ oxidation, leading to relatively higher NH_4^+ concentrations in acidic
452 soils, which causes a dilution effect of the $^{15}NH_4^+$ pool at the initial time (0.5 h) under
453 the $^{15}NH_4NO_3$ treatment. There is increasing evidence that soil pH plays a central role
454 in shaping the communities of active ammonia oxidizers in response to soil conditions.
455 For example, autotrophic nitrification is driven by ammonia-oxidizing bacteria (AOB)
456 rather than ammonia-oxidizing archaea (AOA) in alkaline agricultural soils, while
457 AOA control autotrophic nitrification in acidic agricultural soils (Jiang et al., 2015;
458 Shi et al., 2016).

Heterotrophic nitrification was only observed in the Lishu soil with gross rates of 0.80 mg N $kg^{-1}d^{-1}$. Generally, heterotrophic nitrification is observed in forest and grassland soils which are characterized by higher organic C and N, and is considered to be negligible in agricultural soils (Nelissen et al., 2012; Zhang et al., 2015). This was confirmed by a meta-study highlighting a positive relationship between heterotrophic nitrification and SOC and C/N ratios but showed a negative correlation with soil pH on a global scale (Chen et al., 2015). Based on previous studies, we infer that high SOC, C/N and low pH are crucial factors in controlling heterotrophic nitrification in the Lishu soil.

4.2 Mineralization and NH_4^+-N & NO_3^--N immobilization

Mineralization of labile (M_{Nlab}) and recalcitrant (M_{Nrec}) organic N to inorganic N, total gross mineralization and autotrophic nitrification were significantly positively correlated in all soils except soil from Jinjing with the lowest soil pH, which also showed a higher MIT. Zhang et al. (2018) reviewed the inherent connections between soil N transformations and climate, and showed that gross rates of N mineralization and immobilization (NH_4^+-N & NO_3^--N immobilization) in acidic soils in humid climate zones (mean annual precipitation > 800 mm) were significantly greater than those in semi-humid climate zones (400 mm < mean annual precipitation < 800 mm) and soils in semiarid and arid regions (< 400 mm), which were characterized by a neutral or alkaline pH.

Gross NH_4^+ immobilization rates were very low in most soils except for the Jinjing soil with the lowest pH, which refutes our initial hypothesis that the UK soils

should have the highest capacity for NH_4^+ immobilization due to relatively high soil organic C. So, NH_4^+ immobilization is also controlled by soil pH regardless of origin or organic carbon concentration. However, during the period between 0-0.5 h, it was found that the $\Delta NH_4^+ < \Delta NO_3^-$ in all Chinese soils, but this was reversed in the three UK soils (Fig. S2 a), which indicated net immobilization of NH_4^+ by UK soils was stronger than that in the Chinese soils at the very beginning of addition of NH_4NO_3. This is consistent with work by Silgram and Chambers (2002) who also found temporary net NH_4^+ immobilization in UK soils. This may be because the NH_4^+ concentrations were sufficiently high at the early stage of incubation to support the N demand of both the heterotrophs and the nitrifiers, but as the NH_4^+ concentrations dropped, nitrifiers were more successful in competing for NH_4^+ with immobilizing organisms in later incubation periods (Burger and Jackson, 2003).

Microbial mineralization in Jinjing and Stetchworth soils was significantly higher (p<0.05) than in other soils, which might have been caused by their comparatively high total N and soil organic C contents. This is in line with previous studies showing that gross mineralization rates were positively correlated with soil organic C and total N concentrations, providing the energy and substrates for microbial N cycling processes (Booth et al., 2005; Lang et al., 2010). However, research across ten agricultural soils in the humid zones of China found no significant relationship between gross N mineralization rates and individually measured soil properties including soil organic C, total N and C/N ratio (Zhang et al., 2013a).

Nitrate (NO_3^-) immobilization was low in all of our agricultural soils which has

503 also been shown by others (Shi et al., 2004; Zhang et al., 2012b). Microbial
504 immobilization of NO_3^- requires more energy by comparison to NH_4^+ immobilization
505 and may be suppressed by NH_4^+ even at low concentrations (Templer et al., 2008). In
506 addition, the quantity and quality of soil organic C are also considered as key factors
507 affecting NO_3^- immobilization (Burger and Jackson, 2003; Häbteselassie et al., 2006).
508 Previous studies have reported that microbes need to immobilize inorganic N to
509 satisfy their metabolic needs during decomposition of organic matter with high C/N
510 ratios (Janssen, 1955; Zhang et al., 2013b), explaining why significant NO_3^-
511 immobilization is reported in forest, grassland and organically farmed soils that are
512 characterized by much higher organic C contents and high C/N ratios compared to
513 conventionally farmed agricultural soils (Bradley, 2001; Cheng et al., 2012), which
514 have relatively low organic matter inputs, high mineral N fertilization and regular
515 tillage leading to a decline in the ability to retain N. To restrict NO_3^- accumulation and
516 the associated risk of high N losses, it is important to develop management strategies
517 that promote organic C and N retention in cropland systems.

518 4.3 Model simulation of $NH_4^+$and NO_3^- concentrations and their atom % ^{15}N excess

519 According to the *Ntrace* simulation results, we know that the methods for
520 determining gross soil N transformation rates using the ^{15}N labelling technique require
521 that concentrations and enrichments of the measured N change significantly over the
522 measurement period and that the applied ^{15}N pool is not rapidly consumed (Murphy et
523 al., 2003). It is difficult to accurately determine the gross N transformation rates if the
524 supplied ^{15}N (e.g. $^{15}NH_4^+$) is rapidly depleted (Watson et al., 2000; Stark and Schimel,

525 2001). In this study, we also found that the NH_4^+ concentration and its enrichment in
526 the labelled $^{15}NH_4NO_3$ treatment decreased dramatically after 24 h in the neutral and
527 alkaline soils, due to their high ability to transform NH_4^+ to NO_3^- (Fig. 2). Therefore,
528 for the neutral and alkaline soils with their high ability for ammonia oxidation, we
529 suggest that additional soil sampling should be performed at 6 h after ^{15}N labelling to
530 provide more robust data for model calculations of gross N transformation rates.
531

5. Conclusions

Our results showed that there were no intrinsic differences in soil N transformations and N_2O emissions between Chinese and UK agricultural soils, but that differences mainly resulted from differences in soil properties, specifically pH. This leads to the inference that the dynamics of N transformations are globally affected by similar conditions. Autotrophic nitrification dominated NO_3^- production and was closely related to cumulative N_2O emissions under aerobic conditions, which was significantly regulated by soil pH. This understanding of gross N transformation processes and N_2O emission pathways will be valuable in helping to develop more appropriate N management and N_2O mitigation strategies.

Acknowledgments

This work was supported by the National Natural Science Foundation of China (41830751, 41471190), the Newton Funded N Circle project (Grant Ref. BB/N013484/1) and the Scottish Government's Strategic Research Programme. We greatly appreciate the soil sampling assistance from Chong Zhang, Jianlin Shen, Guozhong Feng, Wei Zhou, Xinsheng Niu, Xiaodong Lv, Shengli Guo and Lijun Li.

Appendix A. Supplementary material

Supplementary tables and figures related to this article can be found in Appendix A.

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Table 1 Some important soil-climatic properties of the top layer (0-20 cm) of ten studied agricultural soils [Mean (SD)]

Parameter	Jinjing	Lishu	Changshu	Quzhou	Zhangye	Changwu	Jinzhong	Boghall	Boxworth	Stetchworth
Latitude	28°33′N	43°17′N	31°32′N	36°52′N	37°96′N	35°12′N	37°32′N	55°52′N	52°14′N	52°13′N
Longitude	113°20′E	124°20′E	120°40′E	115°10′E	102°64′E	107°40′E	112°40′E	3°13′W	0°05′E	0°22′E
Soil type[a]	Haplic Acrisol	Haplic Phaeozems	Hydrgric Anthrosol	Calcaric Cambisol	Calcaric Fluvisal	Calcarid Regsol	Capcic Luvisol	Gleyic Cambisol	Calcaric Cambisols	Chromic Luvisols
Soil texture[b]	Loamy clay	Loamy clay	Silty clay	Sandy loam	Slity loam	Sandy loam	Silty loam	Sandy loam	Clay	Sandy loam
Clay (%)	27	30	26	3	18	22	17	18	45	17
Land use (crop)[c]	Rice-Rice (two harvest a year)	Spring maize monoculture (one harvest a year)	Rice-Wheat (two harvest a year)	Wheat-Maize (two harvest a year)	Spring maize Monoculture (one harvest a year)	Spring maize Monoculture (one harvest a year)	Spring maize Monoculture (one harvest a year)	Cereals-Oilseed Rotation (one harvest a year)	Cereals-Oilseed Rotation (one harvest a year)	Cereals-Oilseed Rotation (one harvest a year)
T (℃)[d]	17.5	6.5	15.0	13.2	7.6	9.3	9.7	8.8	12.0	12.0
P (mm)[e]	1330	650	1038	494	152	584	441	567	550	590
Climate	Subtropical monsoon	Temperate monsoon	Subtropical monsoon	Temperate monsoon	Temperate continental	Temperate continental	Temperate Continental	Temperate marine	Temperate marine	Temperate marine
pH	5.4 (0.0)	6.2 (0.0)	7.1 (0.0)	8.3 (0.0)	8.3 (0.0)	8.6 (0.1)	8.7 (0.1)	6.0 (0.0)	7.7 (0.1)	7.9 (0.0)
SOC (g kg^{-1})[f]	25.5 (0.1)	17.8 (0.1)	27.1 (0.2)	9.1 (0.2)	11.4 (0.8)	8.4(0.2)	8.1 (0.0)	24.2 (3.0)	21.9 (1.0)	27.4 (1.4)
TN (g kg^{-1})[g]	2.5 (0.0)	1.7 (0.1)	2.3 (0.0)	0.9(0.0)	1.0 (0.0)	0.9 (0.0)	0.7 (0.0)	1.8(0.2)	2.4 (0.1)	3.0 (0.2)
C/N	10.1 (0.0)	10.2 (0.1)	11.9 (0.1)	10.1 (0.4)	12.0 (0.0)	9.3 (0.3)	10.9 (0.2)	12.3(0.2)	9.0(0.2)	9.0 (0.2)
NH_4^+-N (mg kg^{-1})	8.03 (0.07)	1.41 (0.17)	0.10 (0.00)	0.14 (0.08)	0.75 (0.07)	0.72 (0.03)	0.58 (0.17)	0.03 (0.01)	4.70 (0.07)	0.98 (0.10)
NO_3^--N (mg kg^{-1})	15.37 (0.64)	43.24 (0.67)	9.02 (0.85)	31.96 (0.11)	35.23 (0.67)	14.31 (0.25)	43.56 (4.23)	62.36 (1.34)	19.88 (2.88)	33.77 (1.13)
NH_4^+/NO_3^-	0.52 (0.02)	0.04 (0.00)	0.01 (0.00)	0.00 (0.00)	0.02 (0.00)	0.05 (0.00)	0.01 (0.00)	0.00 (0.00)	0.24 (0.04)	0.03 (0.00)

[a]Soils are described according to the FAO/UNESO (1988) classification; [b]Soil texture are described according to the international grading standard of soil texture; [c]Rotation means changing the type of crops grown in the field each year (or changing from crops to fallow) and Monoculture means growing only one crop year after year in the same field; [d]T is the 6-year (2011-2016) mean annual temperature; [e]P is the 6-year (2011-2016) mean annual precipitation; [f]SOC is the soil organic carbon; [g]TN is the soil total nitrogen.

Table 2 Gross N transformation rates [Mean (SD) /mg N $kg^{-1}d^{-1}$] based on ^{15}N tracing model

Parameter	Jinjing[a]	Lishu	Changshu	Quzhou	Zhangye	Changwu	Jinzhong	Boghall	Boxworth	Stetchworth
M_{Nlab}	4.45 (1.08)	0.50 (0.11)^{d}	1.77 (0.60)^{d}	0.78 (0.47)^{d}	3.20 (0.14)^{d}	0.42 (0.06)^{d}	2.00 (0.06)^{d}	0.95 (0.09)^{d}	2.44 (0.43)^{d}	3.40 (0.09)^{b}
M_{Nrec}	0.81 (0.85)	0.53 (0.11)^{b}	0.85 (0.53)^{b}	0.83 (0.44)^{b}	0.04 (0.04)^{b}	0.90 (0.10)^{b}	0.02 (0.02)^{b}	0.45 (0.09)^{d}	0.47 (0.42)^{d}	0.03 (0.04)^{d}
$I_{NH4\text{-}Nlab}$	2.78 (0.84)	0.02 (0.01)^{d}	0.00 (0.00)^{d}	0.00 (0.00)^{d}	0.00 (0.00)^{d}	0.00 (0.00)^{d}	0.00 (0.00)^{d}	0.00 (0.00)^{d}	0.00 (0.00)^{d}	0.00 (0.00)^{d}
$I_{NH4\text{-}Nrec}$	1.77 (0.60)	0.02 (0.02)^{d}	0.00 (0.00)^{d}	0.00 (0.00)^{d}	0.00 (0.00)^{d}	0.00 (0.00)^{d}	0.00 (0.00)^{d}	0.00 (0.00)^{d}	0.00 (0.00)^{d}	0.00 (0.00)^{d}
O_{Nrec}	0.00 (0.00)	0.80 (0.09)^{d}	0.00 (0.00)^{b}	0.00 (0.00)^{b}	0.00 (0.00)^{b}	0.00 (0.00)^{b}	0.00 (0.00)^{b}	0.00 (0.00)^{b}	0.00 (0.00)^{b}	0.00 (0.00)^{b}
O_{NH4}	0.25 (0.04)	3.05 (0.05)^{d}	8.06 (0.09)^{d}	11.04 (0.08)^{d}	14.24 (0.20)^{d}	6.94 (0.10)^{d}	14.43 (0.17)^{d}	4.15 (0.04)^{d}	13.26 (0.19)^{d}	11.29 (0.09)^{d}
A_{NH4}	0.02 (0.00)	0.00 (0.00)^{b}	0.00 (0.00)^{b}	0.00 (0.00)^{b}	0.00 (0.00)^{b}	0.00 (0.00)^{b}	0.00 (0.00)^{b}	0.00 (0.00)^{b}	0.00 (0.00)^{b}	0.00 (0.00)^{b}
R_{NH4}	0.00 (0.00)	0.00 (0.00)^{b}	0.00 (0.00)^{b}	0.00 (0.00)^{b}	0.00 (0.00)^{b}	0.00 (0.00)^{b}	0.00 (0.00)^{b}	1.33 (0.03)^{d}	0.00 (0.00)^{b}	0.00 (0.00)^{b}
I_{NO3}	0.00 (0.00)	0.00 (0.00)^{b}	0.07 (0.02)^{d}	0.06 (0.02)^{d}	0.14 (0.03)^{d}	0.04 (0.02)^{d}	0.07 (0.04)^{d}	0.14 (0.08)^{d}	0.09 (0.05)^{d}	0.23 (0.16)^{d}
D_{NO3}	0.00 (0.00)	0.00 (0.00)^{b}	0.00 (0.00)^{b}	0.00 (0.00)^{b}	0.01 (0.00)^{b}	0.01 (0.00)^{b}	0.00 (0.00)^{b}	0.03 (0.00)^{b}	0.03 (0.00)^{b}	0.04 (0.00)^{b}

M_{tot}: M_{Nlab} (mineralization of labile organic N to NH_4^+) and M_{Nrec} (mineralization of recalcitrant organic N to NH_4^+);

I_{NH4tot}: I_{NH4_Nlab} (immobilization of NH_4^+ to N_{lab}) and I_{NH4_Nrec} (immobilization of NH_4^+ to N_{rec});

O_{rec}, oxidation of recalcitrant organic-N to NO_3^- ;

O_{NH4}, oxidation of NH_4^+ to NO_3;

I_{NO3}, immobilization of NO_3^- to recalcitrant organic-N;

D_{NO3}, dissimilatory NO_3^- reduction to NH_4^+;

R_{NH4}, release of adsorbed NH_4^+;

A_{NH4}, adsorption of NH_4^+ on cation exchange sites.

[a]The differences in gross N transformation rates between Jinjing soil and other nine soils were compared based on the comparisons of standard deviations and the 95% confidence intervals to distinguish following three cases:

[b]Standard deviations overlap: the parameters are not different;

[c]Standard deviations do not overlap but 95% confidence intervals overlap, the parameters are not significantly different but show a clear tendency to be different;

[d] 95% confidence intervals do not overlap, parameters are significantly different.

Figure captions

Fig. 1 Measured (*point*) and modeled (*line*) concentrations of the NH_4^+ pool and NO_3^- pool in Jinjing (a), Lishu (b), Changshu (c), Quzhou (d), Zhangye (e), Changwu (f), Jinzhong (g), Boghall (h), Boxworth (i) and Stetchworth (j). Error bars represent standard deviation (n=6), some error bars are covered by symbols due to the low standard deviation values.

Fig. 2 Measured (*point*) and modeled (*line*) atom % ^{15}N excess of the NH_4^+ pool and NO_3^- pool in Jinjing (a and a'), Lishu (b and b'), Changshu (c and c'), Quzhou (d and d'), Zhangye (e and e'), Changwu (f and f'), Jinzhong (g and g'), Boghall (h and h'), Boxworth (i and i') and Stetchworth (j and j'). Error bars represent standard deviation (n=3), some error bars are covered by symbols due to the low standard deviation values.

Fig. 3 Cumulative N_2O-N emission in ten studied soils during the 24 h incubation. Different letters indicate significant differences (p<0.05) among soils. Error bars represent standard deviation (n=3).

Fig. 4 Relationship between soil pH and total gross mineralization rate (a), gross autotrophic nitrification rate (b) and cumulative N_2O-N emission (c). In Fig. 4c, the linear relationship between pH and cumulative N_2O emission does not include Changshu soil. Error bars represent standard deviation (n=3). Some error bars are covered by symbols due to the low standard deviation values.

Fig. 5 Relationship between cumulative N_2O-N emission and total gross mineralization rate (a), autotrophic nitrification rate (b) in ten studied soils. In Fig. 5b, the linear relationship between cumulative N_2O-N emission and autotrophic nitrification rate does not include Changshu soil. Error bars represent standard deviation (n=3). Some error bars are covered by symbols due to the low standard deviation values.

Fig. 6 Relationship between total gross mineralization rate and autotrophic nitrification rate excepting for Jinjing soil. Error bars represent standard deviation (n=3). Some error bars are covered by symbols due to the low standard deviation values.

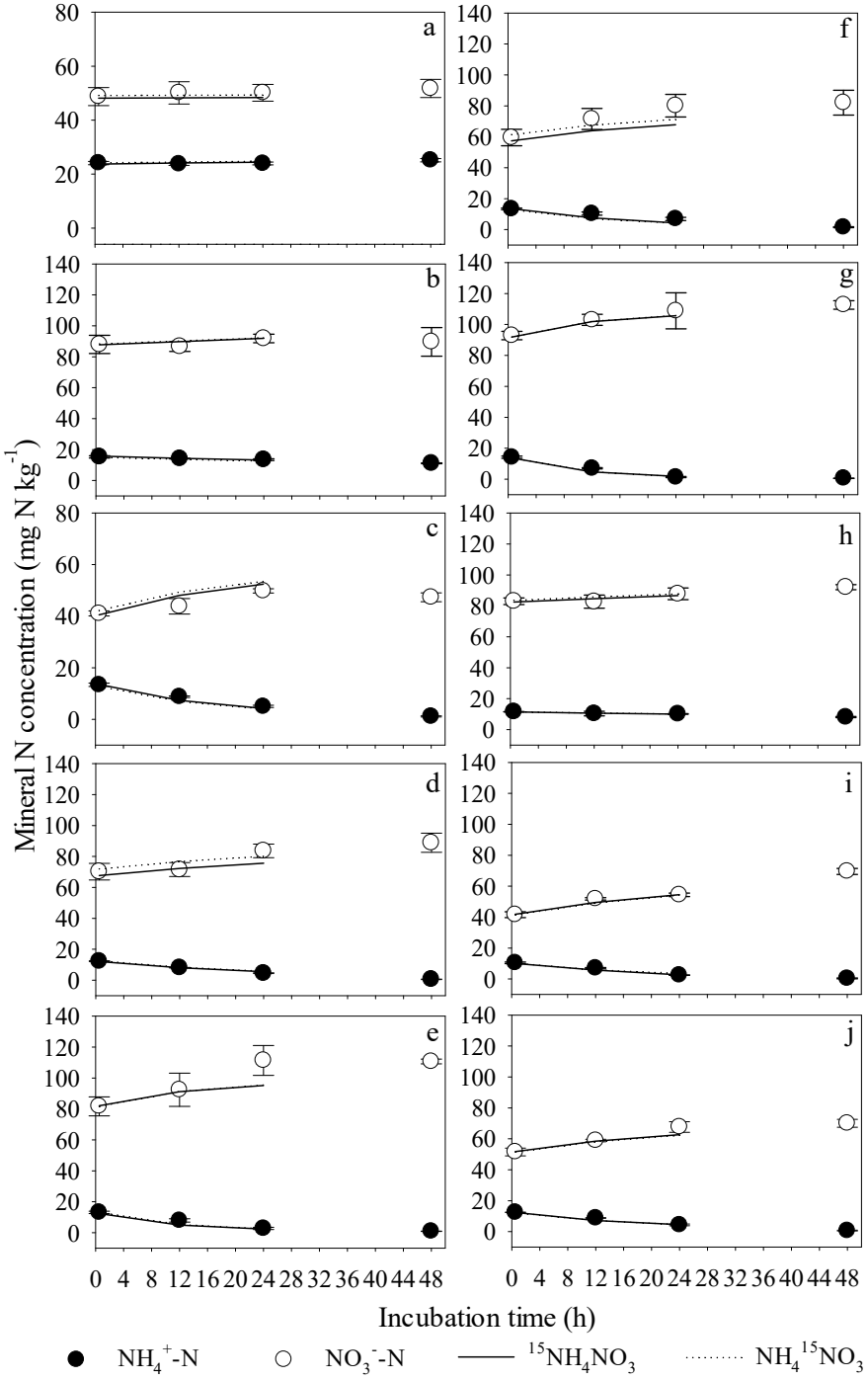
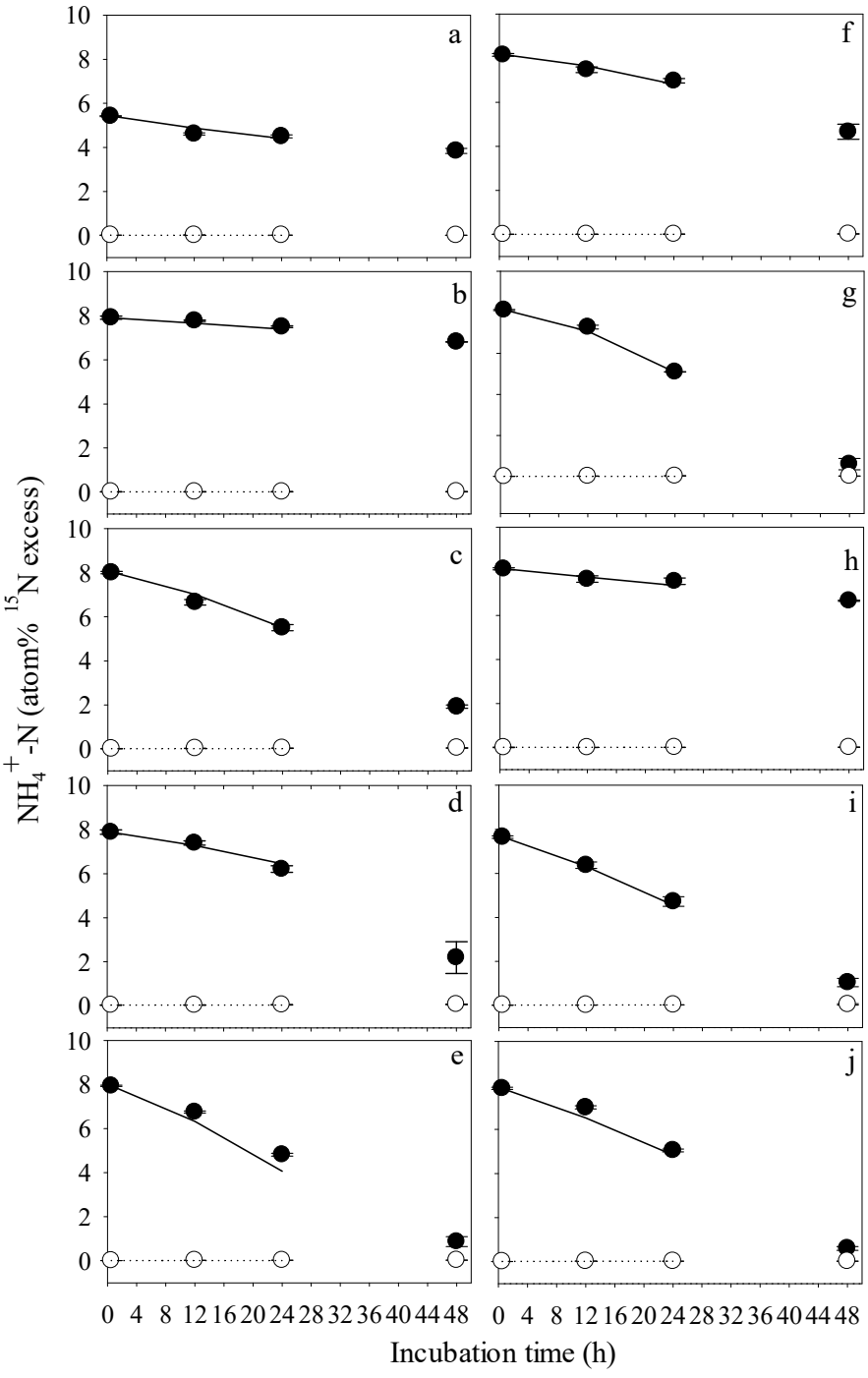

a
b
c
d
e
f
g
h
i
j
Mineral N concentration (mg N kg-1)
Incubation time (h)
0 4 8 12 16 20 24 28 32 36 40 44 48
NH4+-N
NO3--N
15NH4NO3
NH415NO3

Fig. 1 (Zhu GD)


a
b
c
d
e
f
g
h
i
j
NH_4^+-N (atom% ^{15}N excess)
Incubation time (h)
0 4 8 12 16 20 24 28 32 36 40 44 48
0 2 4 6 8 10

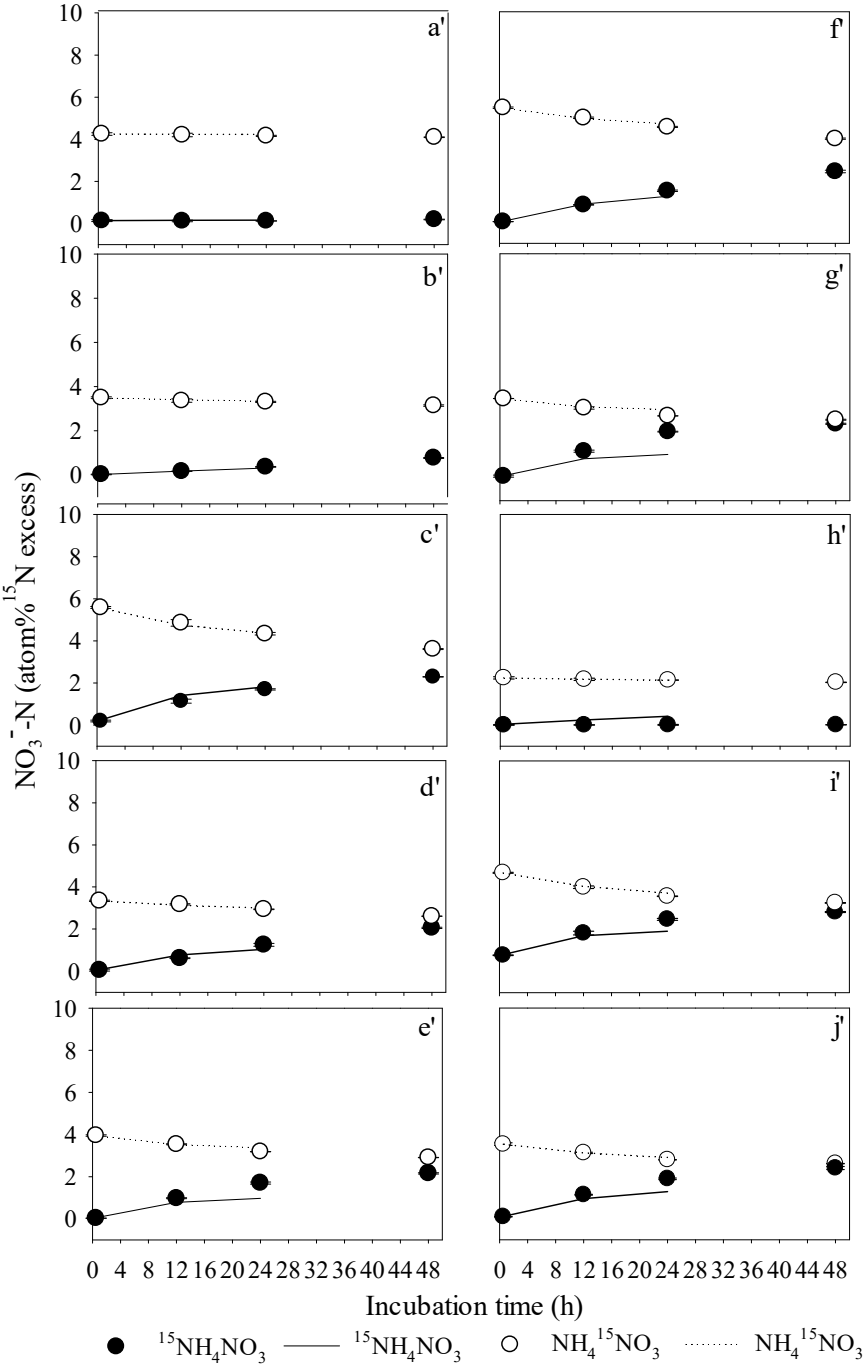

a'
b'
c'
d'
e'
f'
g'
h'
i'
j'
NO_3^--N (atom% ^{15}N excess)
Incubation time (h)
$^{15}NH_4NO_3$
$^{15}NH_4NO_3$
$NH_4{}^{15}NO_3$
$NH_4{}^{15}NO_3$

Fig. 2 (Zhu GD)

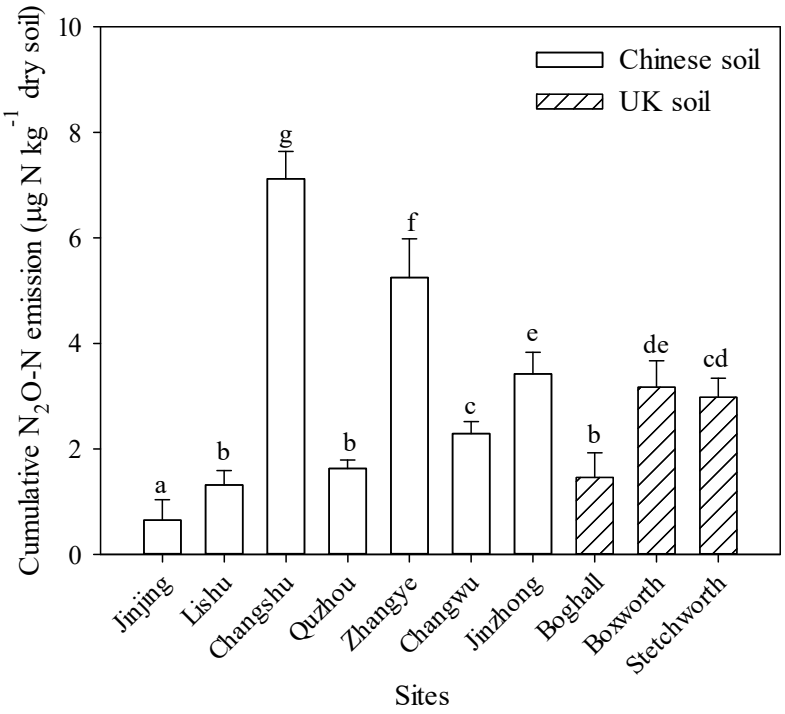

Cumulative N_2O-N emission (μg N kg^{-1} dry soil)
10
8
6
4
2
0
Chinese soil
UK soil
a
b
g
b
f
c
e
b
de
cd
Jinjing
Lishu
Changshu
Quzhou
Zhangye
Changwu
Jinzhong
Boghall
Boxworth
Stetchworth
Sites

Fig. 3 (Zhu GD)

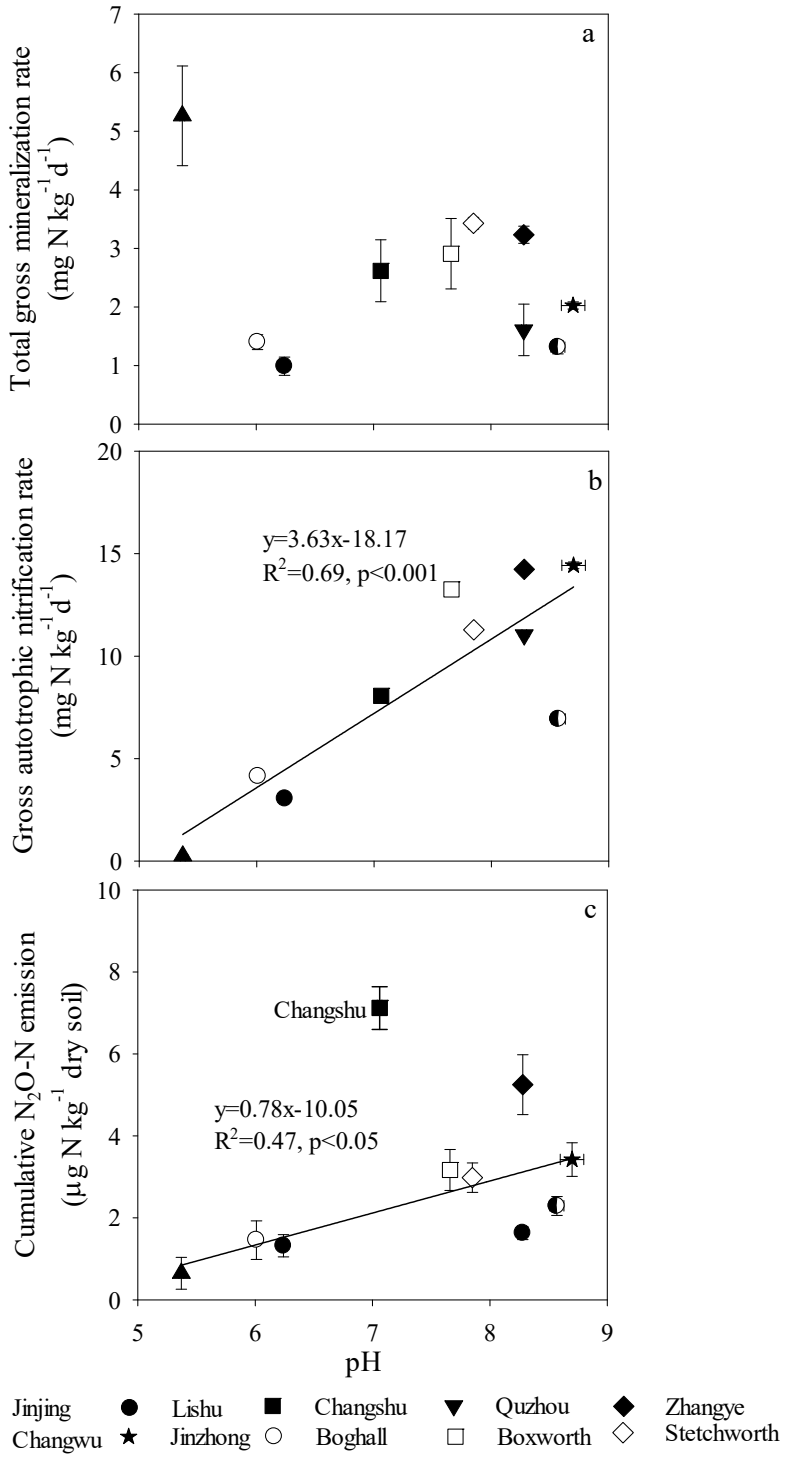

a
Total gross mineralization rate (mg N kg-1 d-1)
b
Gross autotrophic nitrification rate (mg N kg-1 d-1)
y=3.63x-18.17
R2=0.69, p<0.001
c
Cumulative N2O-N emission (µg N kg-1 dry soil)
Changshu
y=0.78x-10.05
R2=0.47, p<0.05
pH
Jinjing
Lishu
Changshu
Quzhou
Zhangye
Changwu
Jinzhong
Boghall
Boxworth
Stetchworth

Fig. 4 (Zhu GD)

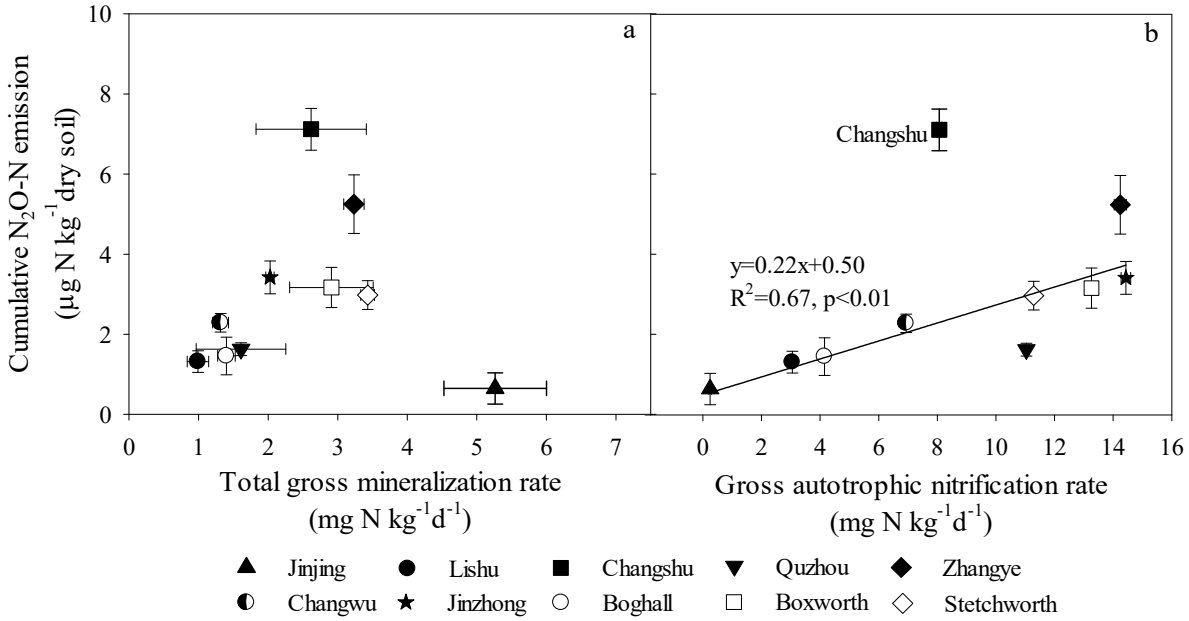
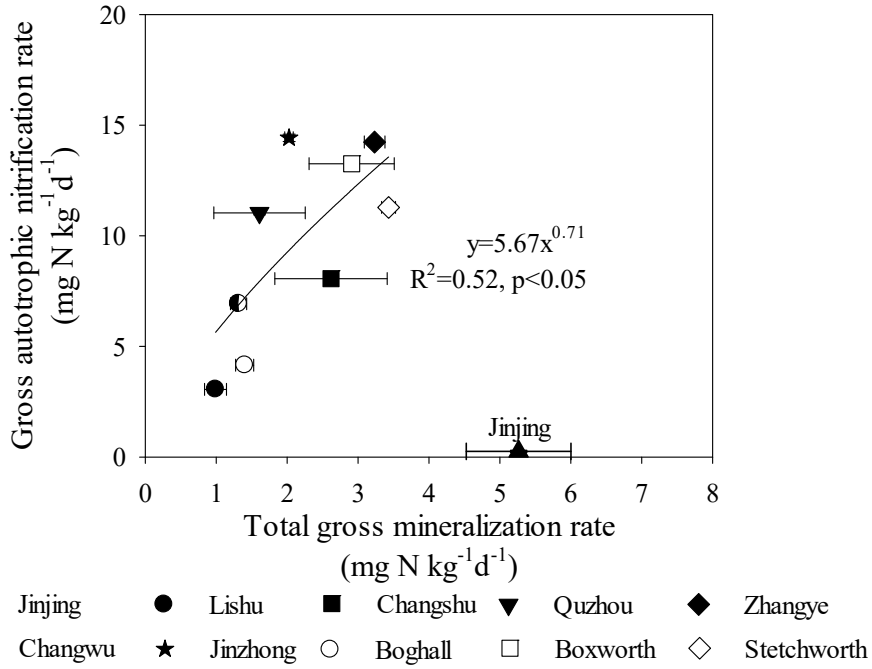

a
b
Cumulative N_2O-N emission
(μg N kg^{-1} dry soil)
Total gross mineralization rate
(mg N $kg^{-1}d^{-1}$)
Gross autotrophic nitrification rate
(mg N $kg^{-1}d^{-1}$)
Changshu
y=0.22x+0.50
R^2=0.67, p<0.01
Jinjing
Lishu
Changshu
Quzhou
Zhangye
Changwu
Jinzhong
Boghall
Boxworth
Stetchworth

Fig. 5 (Zhu GD)

39



40

41 **Fig. 6 (Zhu GD)**

Table S1 NH_4^+ and NO_3^- concentrations [Mean (SD) / mg N kg^{-1}] at different sampling times and net mineralization and nitrification rate (mg N $kg^{-1}d^{-1}$) during different incubation periods in ten studied soils.

	Initial NH_4^+ in soil	Expected NH_4^+ after added NH_4NO_3 in zero time	NH_4^+ at 0.5h	NH_4^+ at 12h	NH_4^+ at 24h	NH_4^+ at 48h	Initial NO_3^- in soil	Expected NO_3^- after added NH_4NO_3 in zero time	NO_3^- at 0.5h	NO_3^- at 12 h	NO_3^- at 24 h	NO_3^- at 48 h
			A	B	C	D			E	F	G	H
Jinjing	8.03	28.03	24.28	23.78	23.94	25.94	15.37	35.37	48.73	50.57	50.15	51.73
	(0.07)	(0.07)	(0.65)	(0.55)	(0.54)	(0.54)	(0.64)	(0.64)	(3.33)	(3.46)	(3.11)	(3.40)
Lishu	1.41	21.41	15.34	14.18	13.55	11.18	43.24	63.24	87.99	86.61	91.89	89.67
	(0.17)	(0.17)	(0.70)	(0.25)	(0.54)	(0.25)	(0.67)	(0.67)	(5.84)	(3.13)	(2.77)	(9.22)
Changshu	0.10	20.10	13.40	8.78	5.03	1.15	9.02	29.02	41.07	43.86	49.82	47.35
	(0.00)	(0.00)	(0.65)	(0.27)	(0.42)	(0.22)	(0.85)	(0.85)	(1.00)	(3.04)	(0.84)	(1.69)
Quzhou	0.14	20.14	12.27	8.08	4.47	0.48	31.96	51.96	70.14	74.55	83.71	88.90
	(0.08)	(0.08)	(0.23)	(0.19)	(0.23)	(0.12)	(0.11)	(0.11)	(5.46)	(0.87)	(4.38)	(6.17)
Zhangye	0.75	20.75	13.09	7.84	2.38	0.89	35.23	55.23	81.65	86.83	111.36	110.66
	(0.07)	(0.07)	(0.73)	(1.08)	(0.77)	(0.08)	(0.67)	(0.67)	(6.14)	(8.36)	(9.68)	(1.52)
Changwu	0.72	20.72	13.47	10.43	6.97	1.63	14.31	34.31	59.58	69.52	80.20	82.11
	(0.03)	(0.03)	(0.47)	(1.04)	(1.01)	(0.35)	(0.25)	(0.25)	(5.30)	(5.13)	(7.27)	(8.02)
Jinzhong	0.58	20.58	14.26	7.13	1.42	0.60	43.56	63.56	92.87	103.04	115.86	112.70
	(0.17)	(0.17)	(0.77)	(0.38)	(0.29)	(0.06)	(4.23)	(4.23)	(2.74)	(3.57)	(11.67)	(2.81)
Boghall	0.03	20.03	11.58	10.95	10.13	8.12	62.36	82.36	83.03	82.85	87.81	92.18
	(0.01)	(0.01)	(0.12)	(0.26)	(0.22)	(0.40)	(1.34)	(1.34)	(2.12)	(1.66)	(3.80)	(1.63)
Boxworth	4.70	24.70	10.44	6.97	2.50	0.37	19.88	39.88	41.57	51.75	54.65	69.57
	(0.07)	(0.07)	(0.46)	(0.42)	(0.02)	(0.25)	(2.88)	(2.88)	(1.86)	(0.76)	(1.12)	(2.03)
Stetchworth	0.98	20.98	12.43	8.81	4.33	0.66	33.77	53.77	51.48	58.86	67.62	70.03
	(0.10)	(0.10)	(0.10)	(0.21)	(0.53)	(0.23)	(1.13)	(1.13)	(2.48)	(0.85)	(3.37)	(2.57)

ΔNH_4^+ (0.5h-12h)	ΔNH_4^+ (12h-24h)	ΔNH_4^+ (24h-48 h)	ΔNH_4^+ (0.5h-24h)	ΔNH_4^+ (0.5h-48 h)	ΔNO_3^- (0.5h-12h)	ΔNO_3^- (12h-24h)	ΔNO_3^- (24h-48 h)	ΔNO_3^- (0.5h-24h)	ΔNO_3^- (0.5h-48 h)	Net mineralization (0.5h-12h)	Net mineralization (0.5h-24h)	Net mineralization (0.5h-48h)	Net nitrification (0.5h-12h)	Net nitrification (0.5h-24h)	Net nitrification (0.5h-48h)
A-B	B-C	C-D	A-C	A-D	E-F	F-G	G-H	E-G	E-H						
0.50 (0.42)	-0.25 (0.58)	-1.21 (0.33)	0.38 (0.49)	-0.88 (0.52)	-1.00 (0.66)	-1.5 (3.22)	0.1 (2.32)	-1.42 (2.18)	-1.60 (2.79)	2.78 (3.65)	2.30 (1.81)	2.94 (1.49)	6.26 (1.38)	1.28 (0.90)	2.59 (1.74)
1.17 (0.61)	0.85 (0.34)	2.38 (0.64)	1.79 (0.98)	4.17 (0.82)	3.30 (1.52)	-5.27 (1.71)	-0.5 (1.17)	-2.69 (1.28)	-8.84 (3.94)	-7.87 (2.66)	1.84 (1.67)	3.04 (1.58)	-8.44 (0.71)	2.75 (1.31)	4.47 (1.90)
4.63 (0.44)	3.75 (0.35)	3.88 (0.45)	8.38 (0.34)	12.25 (0.68)	-4.50 (0.20)	-5.96 (3.01)	1.58 (0.29)	-8.75 (0.60)	-6.28 (0.93)	-3.30 (4.38)	0.57 (0.37)	-3.01 (0.74)	9.39 (0.43)	8.94 (0.68)	3.17 (0.47)
4.19 (0.34)	3.61 (0.37)	3.99 (0.29)	7.80 (0.16)	11.79 (0.28)	-4.85 (2.31)	-9.16 (3.84)	-4.08 (2.38)	-13.57 (2.17)	-19.09 (4.21)	0.44 (3.07)	5.21 (1.60)	3.70 (2.27)	16.30 (5.27)	13.85 (2.21)	9.64 (2.13)
5.25 (0.88)	5.17 (0.85)	1.78 (0.76)	10.42 (0.98)	12.20 (0.78)	-15.06 (4.21)	-8.37 (4.40)	-2.97 (1.90)	-29.71 (6.99)	-29.01 (6.04)	-0.15 (5.32)	19.42 (2.41)	9.33 (2.41)	31.42 (8.80)	28.18 (5.32)	14.66 (3.05)
3.04 (1.21)	3.46 (1.49)	5.33 (0.82)	6.5 (1.11)	11.83 (0.61)	-7.97 (4.15)	-7.0 (3.13)	-5.90 (1.55)	-15.25 (2.30)	-22.04 (0.31)	13.31 (8.76)	8.56 (3.06)	5.30 (0.27)	19.31 (7.24)	15.58 (2.34)	11.14 (0.15)
7.13 (0.88)	5.71 (0.29)	0.83 (0.34)	12.84 (0.82)	13.66 (0.80)	-8.12 (1.09)	-12.82 (1.93)	6.89 (0.98)	-22.99 (3.32)	-19.84 (5.05)	6.35 (2.61)	11.68 (3.01)	4.18 (1.43)	20.61 (2.29)	23.48 (3.39)	10.94 (1.37)
0.79 (0.09)	0.76 (0.13)	2.01 (0.67)	1.45 (0.25)	3.46 (0.47)	1.12 (2.12)	-3.57 (2.22)	-2.54 (1.40)	-6.17 (2.22)	-9.16 (2.21)	-1.61 (1.68)	4.90 (2.40)	3.24 (0.76)	-2.95 (4.58)	6.30 (2.26)	4.63 (1.12)
3.49 (0.65)	5.13 (1.62)	1.90 (0.85)	7.94 (0.47)	9.84 (1.07)	-9.95 (1.58)	-3.34 (2.44)	-16.12 (2.01)	-11.98 (1.12)	-28.0 (1.27)	13.49 (2.61)	4.13 (1.41)	9.18 (0.96)	20.77 (3.30)	13.36 (2.93)	14.15 (0.64)
3.62 (0.19)	4.48 (0.47)	3.95 (0.53)	8.10 (0.44)	12.04 (0.46)	-7.38 (1.83)	-7.65 (2.40)	-2.26 (0.98)	-14.97 (2.83)	-18.55 (3.97)	6.57 (2.17)	8.22 (3.71)	3.92 (1.39)	14.11 (2.38)	15.29 (2.90)	9.37 (2.00)

Figure S1 Map showing the sampling sites in China (a) and UK (b).

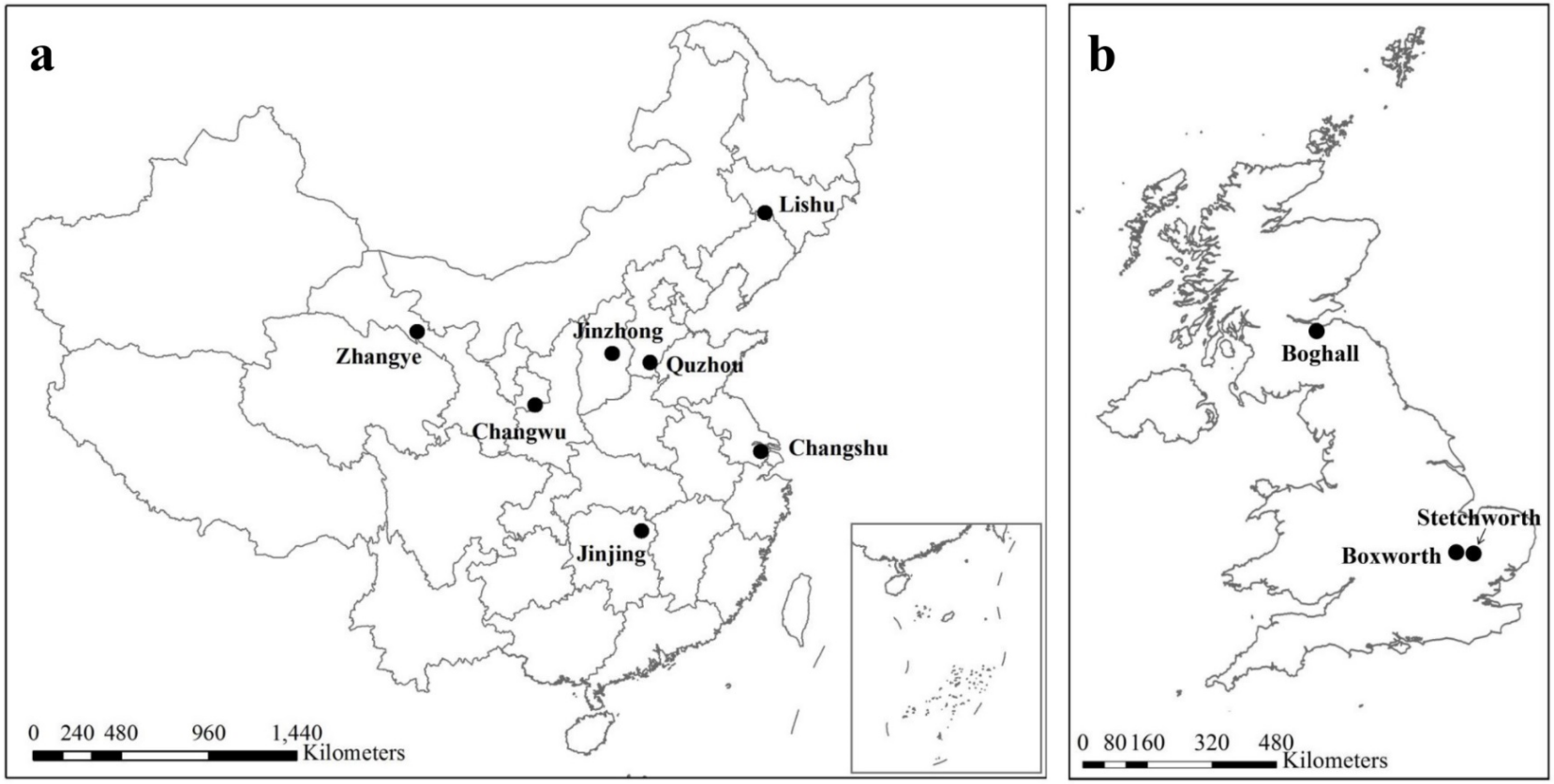


Figure S2 Relationships between the change of NH_4^+ concentration (ΔNH_4^+) and change of NO_3^- concentration (ΔNO_3^-) during 0h-0.5h (a), 0.5h-12h (b), 12h-24h (c), 24h-48h (d) and 0.5h-24h (e) and 0.5h-48h (f). Error bars represent standard deviation (n=6), some error bars are covered by symbols due to the low standard deviation values.

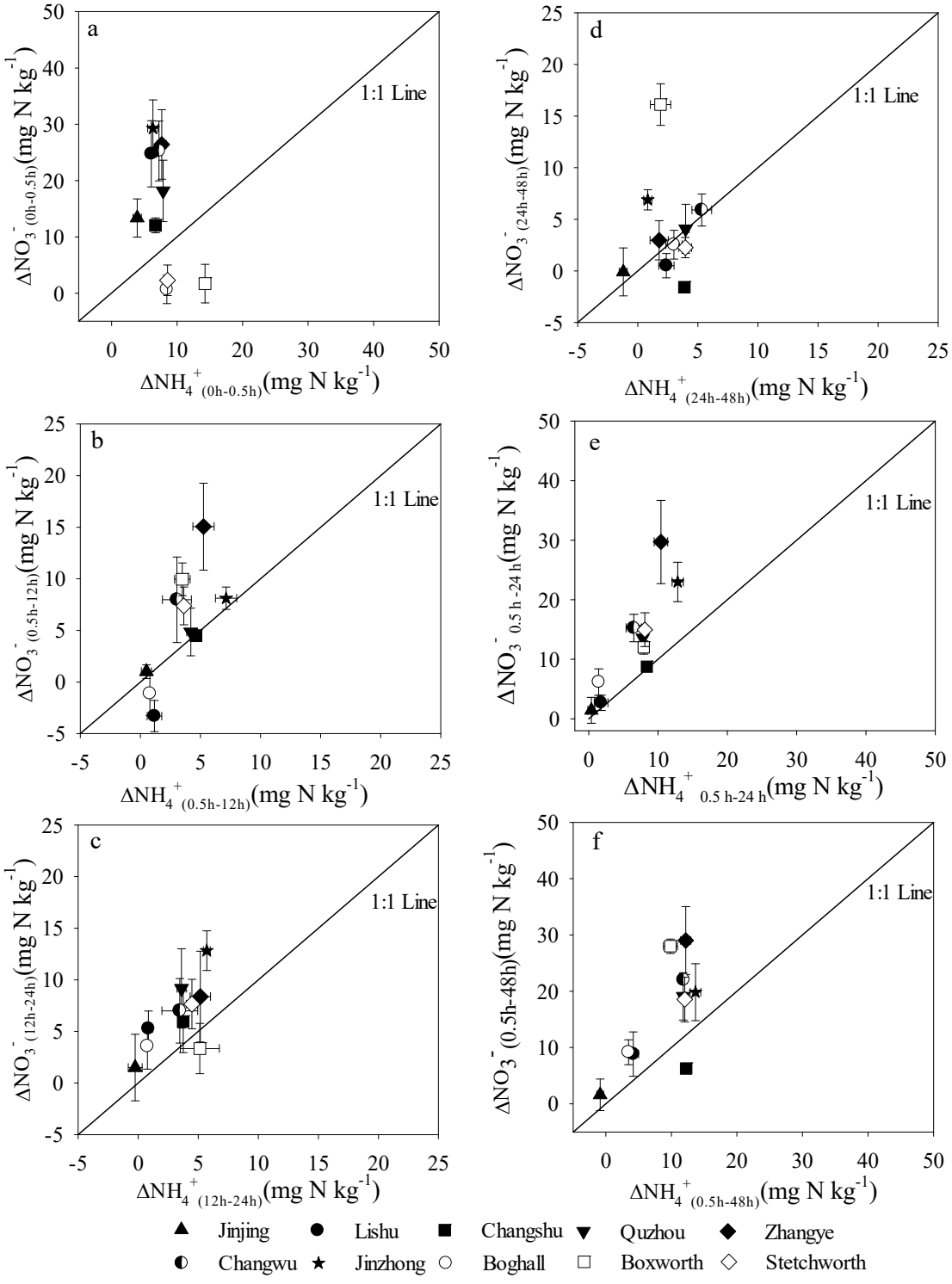


Figure S3 Relationship between initial NH_4^+ concentration and atom% ^{15}N excess of the NH_4^+ pool (0.5 h) in $^{15}NH_4NO_3$ treatment (a), and initial NO_3^- concentration and atom% ^{15}N excess of the NO_3^- pool (0.5 h) in $NH_4{}^{15}NO_3$ treatment (b) in ten studied soils. Error bars represent standard deviation (n=3), some error bars are covered by symbols due to the low standard deviation values.

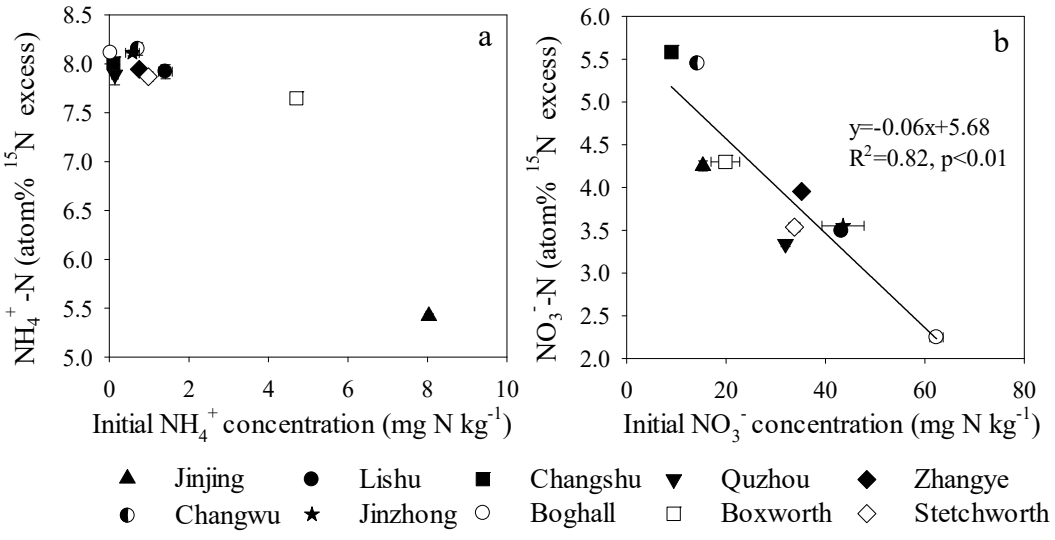


Figure S4 Relationship between NH_4^+-N concentration at 24h and the change of atom % ^{15}N excess of the NH_4^+ pool in the $^{15}NH_4NO_3$ treatment during 24h-48h incubation.

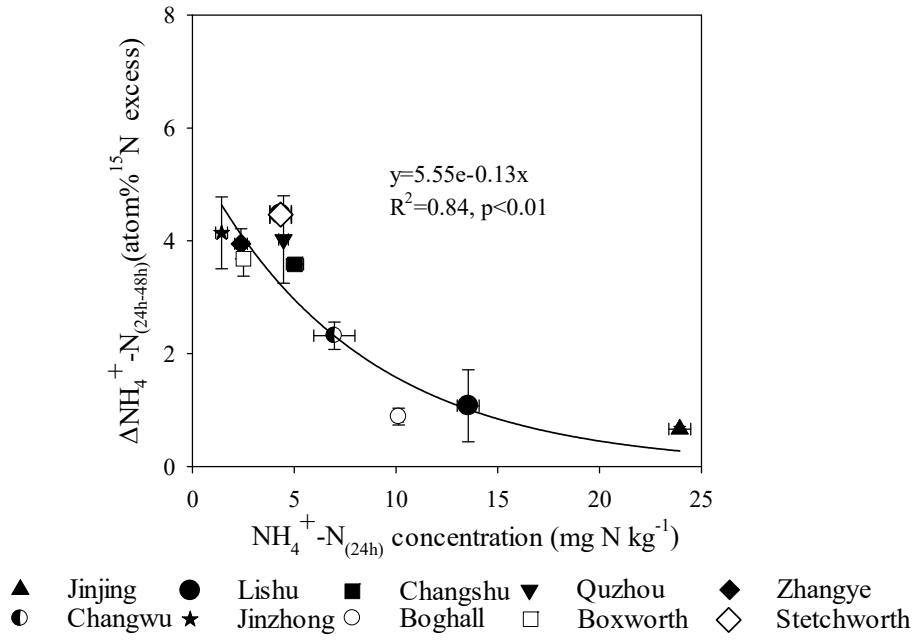


Figure S5 N_2O flux (a), and cumulative N_2O emission (b) in the ten studied soils during the 48h incubation. Error bars represent standard deviation (n=3). Some error bars in Fig. S5a are covered by symbols due to the low standard deviation values. In Fig.S5b, different letters indicate significant differences ($p < 0.05$) among soils.

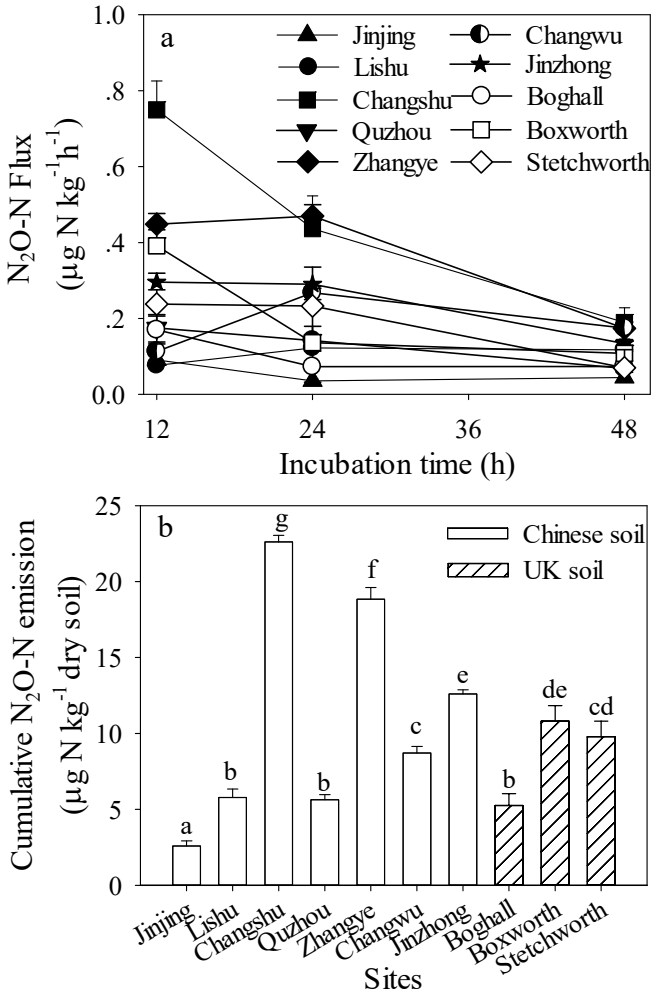


Figure S6 Relationship between total gross mineralization rate and net mineralization rate during the 0.5h-12h (a) and 0.5h-24h (b), and total gross nitrification rate and net nitrification rate during the 0.5h-12h (c) and 0.5h-24h (d) incubation time in ten studied soils. Error bars represent standard deviation (horizontal error, n=3; vertical error, n=6), some error bars are covered by symbols due to the low standard deviation values.

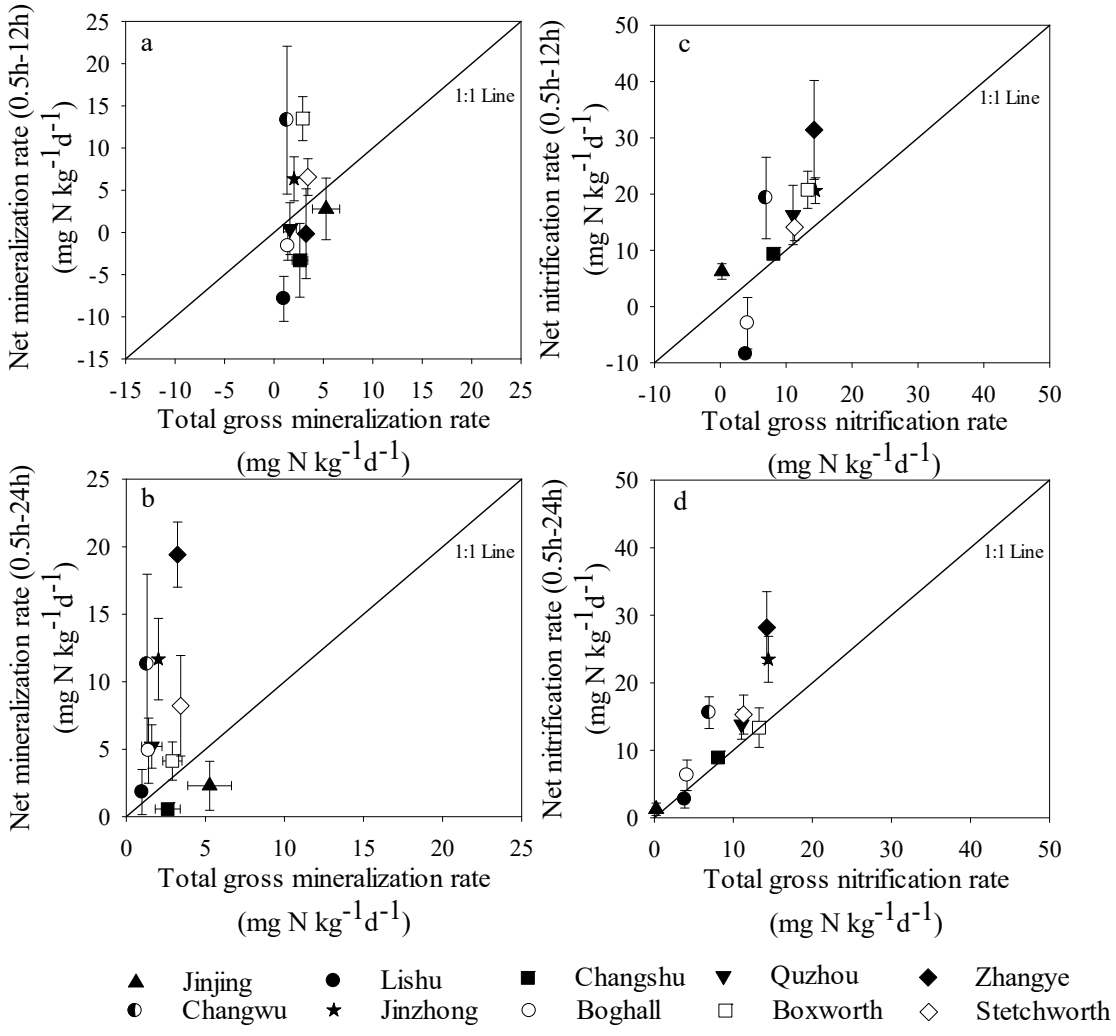


Figure S7 Relationship between total gross mineralization rate and SOC (a), TN (b) and C/N (c); between gross autotrophic nitrification rate and SOC (d), TN (e) and C/N (f) in ten studied soils. Error bars represent standard deviation (n=3), some error bars are covered by symbols due to the low standard deviation values.